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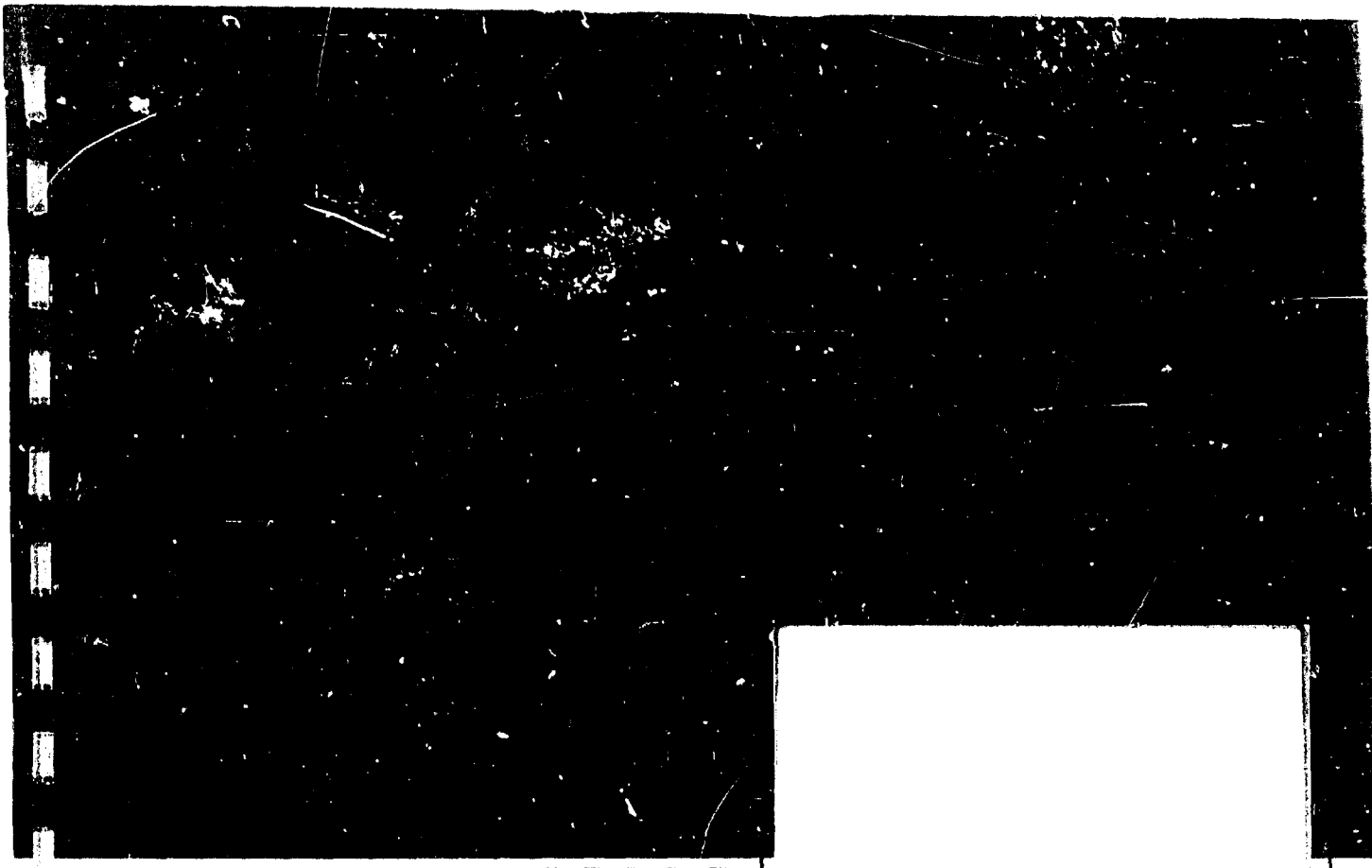
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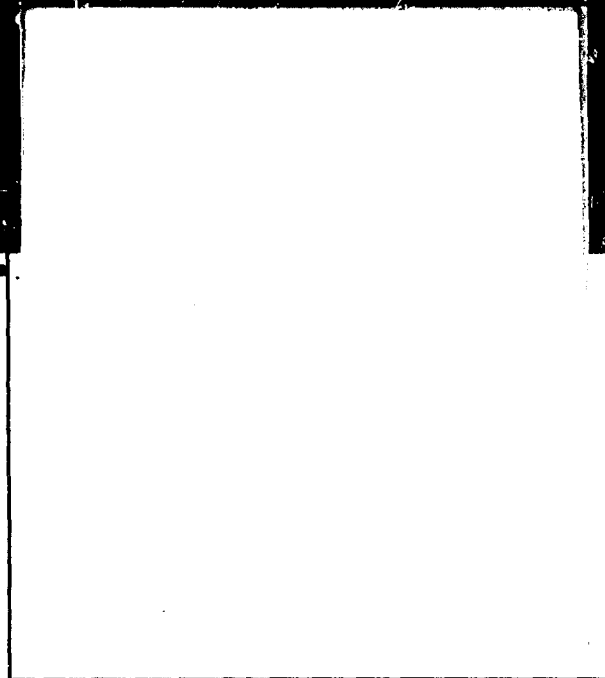
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MATERIALS RESEARCH AND DEVELOPMENT

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This report will be presented at a colloquium on "PHASE STABILITY IN METALS AND ALLOYS" to be held at Geneva, Switzerland in March, 1966 under sponsorship of Battelle Memorial Institute.

THE LATTICE STABILITY OF
METALS IV, TRANSITION
ELEMENTS OF THE SECOND AND
THIRD PERIODS

by

Larry Kaufman

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November 1965

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ABSTRACT

The enthalpy and entropy differences between the b.c.c., h.c.p. and f.c.c. structures have been estimated for transition elements of the second and third periods. Continuous curves depicting these differences as a function of electron number in the zirconium through cadmium and hafnium through gold series are presented. The difference parameters have been used to compute thirty binary phase diagrams between elements in the niobium/tantalum through palladium/platinum groups. These binary diagrams, which are restricted to cases where the end members exhibit differing stable crystal structures and miscibility gaps are absent, have been calculated on the basis of ideal liquid, b.c.c., h.c.p. and f.c.c. phases. The latter simplification has been chosen in order to afford the clearest possible comparison of computed and observed phase diagrams.

The Lattice Stability of Metals IV, Transition
Elements of the Second and Third Periods^{*}

by

Larry Kaufman^{**}

I. INTRODUCTION

Previous papers in this series have dealt with the relative stability of the body centered cubic, hexagonal, close packed and face centered cubic configuration of metals. In particular, it was shown (1)[†] that the difference in free energy between crystallographic forms of a metal made an important contribution in controlling alloying behavior, phase transformations, and stacking fault energy of the metal over a wide range of temperature and pressure. Moreover, the difference in free energy between crystal forms, a quantity often much smaller than present errors in cohesive energy calculations, was shown to be relevant in interpreting specific kinds of activity data (1). In view of present difficulties in making estimates of the free energy difference and its temperature dependence from quantum mechanical principles, a thermodynamic approach based on empirical data has been employed. In this way, lattice stability information for titanium (1), zirconium (1), zinc (2), copper (2), silver (2) and iron (3) have been obtained. In addition, high pressure studies (4, 5) have been employed as a means for deriving lattice stability information.

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** ManLabs, Inc., Cambridge, Massachusetts

† Underscored numbers in parentheses refer to references given at the end of this paper.

Recently, an analysis of the iron-ruthenium system was performed using the regular solution model and assuming that the lattice stability of h.c.p., f.c.c. and b.c.c. ruthenium was similar to nonmagnetic iron. The computed phase diagram for the Fe-Ru system as well as the description of f.c.c. \rightleftharpoons b.c.c. and f.c.c. \rightleftharpoons h.c.p. martensitic transformations at one atmosphere and high pressure resulting from these assumptions was found to agree materially with observation (6). In view of this finding, and the fact that transition elements of the second and third long period exhibit a more regular pattern of crystal structure stability than their counterparts in the first period, it seems worthwhile to consider an analysis of the lattice stability of these elements. The absence of magnetic contributions to the second and third period counterparts of chromium, manganese and iron is undoubtedly the cause of the more regular progression from b.c.c. to h.c.p. to f.c.c. stability across the transition series. In order to make a first approximation to this problem, it has been assumed that second and third row counterparts i. e., Nb and Ta, Ru and Os or Pd and Pt can be characterized by identical lattice stability parameters. Although this need not be true, it is introduced as a simplifying assumption.

On this basis, curves of the enthalpy and entropy differences can be estimated using the available results for h.c.p. and b.c.c. Zr (1); h.c.p., b.c.c. and f.c.c. Ru (6); f.c.c. and b.c.c. Ag (2) and h.c.p., b.c.c. and f.c.c. Cd. The latter values were assumed similar to those for Zn (2). With these values as fixed points, a series of curves can be drawn which are limited by observed stability relations i. e., Re is h.c.p. up to its melting point. In order to obtain better resolution, it became necessary to consider the binary phase diagrams between elements of the second and third row transition

elements. These binary diagrams were chosen for those cases where the component elements have differing stable crystal structures i.e., W-Ir or Mo-Re. In order to use these diagrams to advantage, the rather drastic assumption was made that the h.c.p., b.c.c., f.c.c. and liquid phases could be treated as ideal solutions. As a consequence, systems exhibiting miscibility gaps were eliminated from consideration. Surprisingly enough, as detailed below, a consistent series of lattice stability parameters resulted. These parameters have been used to compute thirty binary phase diagrams which agree quite well with observations. As a consequence, it is concluded that the aforementioned lattice stability values constitute a good first approximation to defining the relative stability of these elements.

II. ENTHALPY AND ENTROPY DIFFERENCES BETWEEN THE H. C. P., B. C. C., AND F. C. C. FORMS AND THE COMPUTATIONS OF BINARY PHASE EQUILIBRIA

The lattice stability of the b. c. c. (β), h. c. p. (ϵ), and f. c. c. (α), configuration of the second and third row transition elements is depicted in Figures 1 and 2.* As indicated earlier, second and third row counterparts (i. e., Nb/Ta or Rh/Ir) are assumed to have identical lattice stabilities. This need not be true as seen in the Zr/Hf case where the enthalpy difference $\Delta H^{\beta \rightarrow \epsilon}$ is more negative for hafnium than for zirconium. However, the assumption of equal parameters has been introduced for simplicity. It will be modified in later studies when the stability of these phases are considered in greater detail.

The equations for binary equilibria between two regular solution phases, ϵ and L, at a fixed temperature where the composition of element B at the $\epsilon/\epsilon + L$ boundary is x_ϵ and the composition at the L + ϵ/L boundary is x_L can be expressed as (1)

$$\Delta F_A^{\epsilon \rightarrow L} + RT \ln \frac{1-x_L}{1-x_\epsilon} = x_\epsilon^2 E - x_L^2 L \quad (1)$$

$$\Delta F_B^{\epsilon \rightarrow L} + RT \ln \frac{x_L}{x_\epsilon} = (1-x_\epsilon)^2 E - (1-x_L)^2 L \quad (2)$$

In Eqs. (1) and (2) the free energy differences $\Delta F_A^{\epsilon \rightarrow L}$ and $\Delta F_B^{\epsilon \rightarrow L}$ represent the differences between the hexagonal and liquid form of elements A and B at the temperature in question. The regular solution interaction parameters for the ϵ and L phases are E and L respectively. If E and L are equal to zero

*These entropy and enthalpy differences refer to high temperatures far from the range influenced by Third Law constraints.

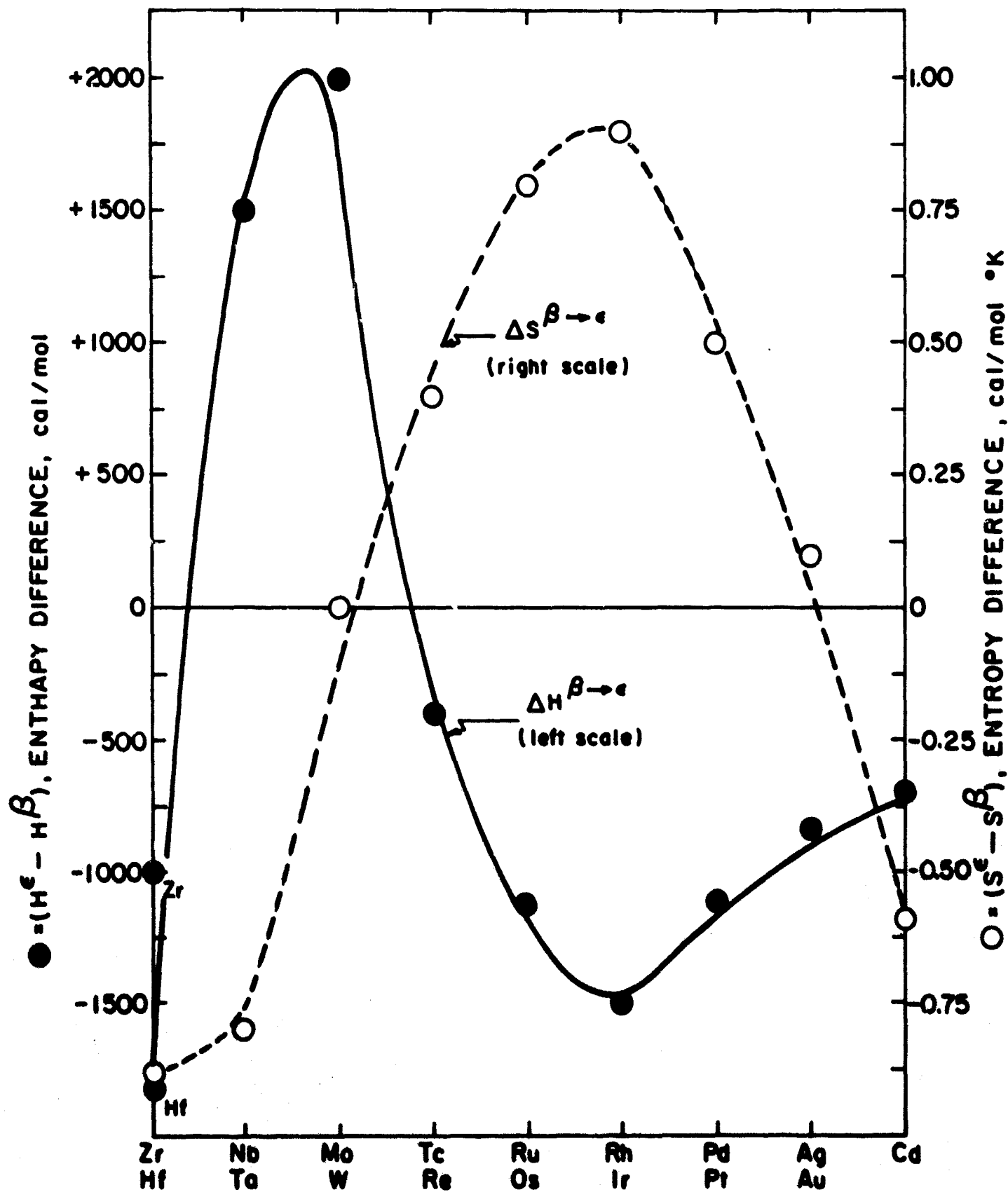


Figure 1. Enthalpy and Entropy Differences Between the h.c.p. (ϵ) and b.c.c. (β) Forms of the Transition Metals.

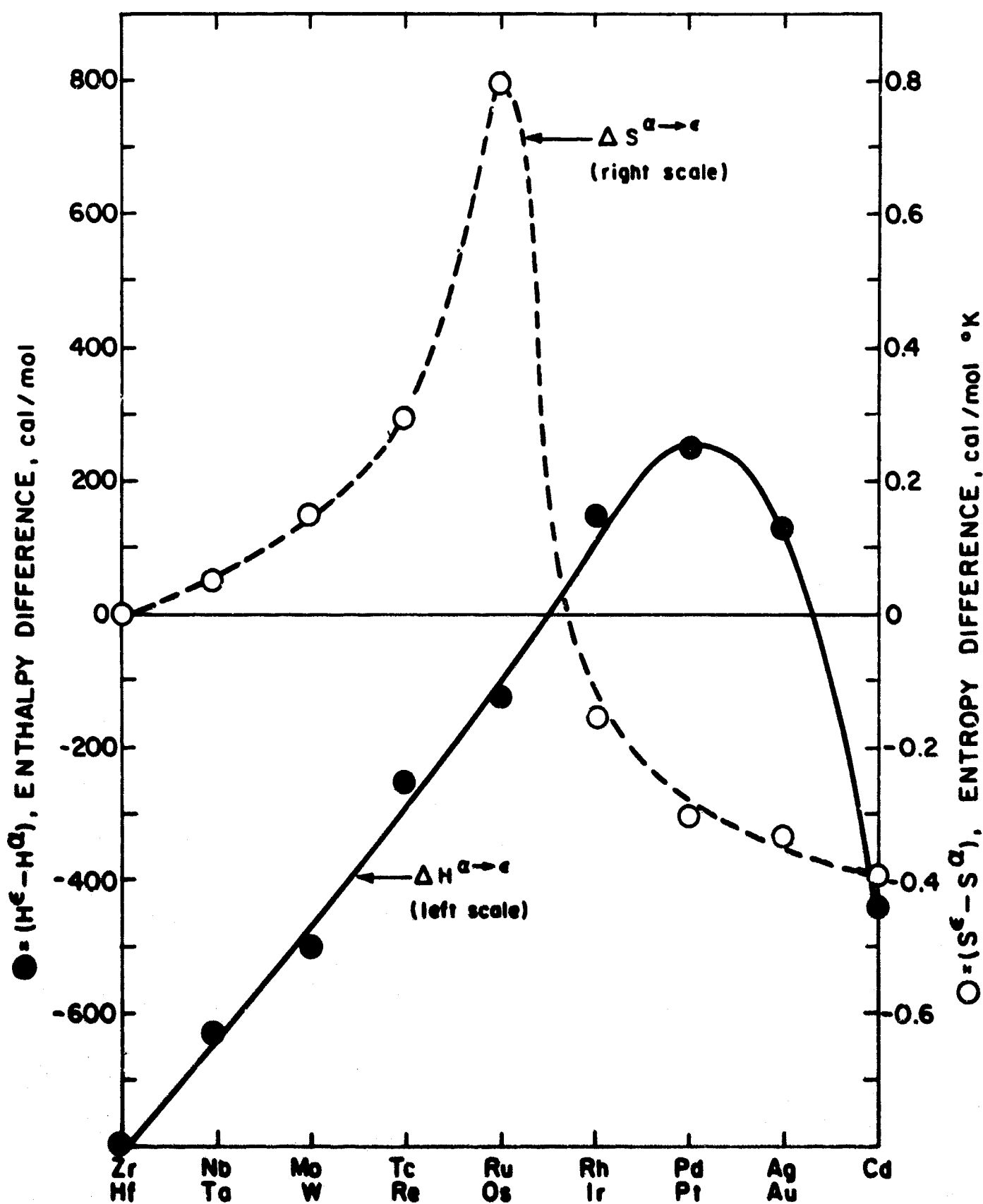


Figure 2. Enthalpy and Entropy Differences Between the h.c.p. (ϵ) and f.c.c. (α) Forms of the Transition Metals.

then the solutions are ideal and the phase boundaries x_ϵ and x_L depend only on the free energy terms for the pure metals. Analogues of Eqs. (1) and (2) can be written for β/L , α/L , ϵ/β , ϵ/α and α/β equilibrium. These equations, when coupled with the information contained in Figures 1 and 2 permit direct computation of ideal solution phase diagrams. The only additional information required is the melting point, \bar{T} , and the entropy of fusion of the stable form elements concerned. In the present case, the entropy of melting was taken to be 2.0 cal/mol $^\circ\text{K}$ in each of the following instances where experimental data is not available: niobium ($\bar{T}^\beta = 2740^\circ\text{K}$), tantalum ($\bar{T}^\beta = 3270^\circ\text{K}$), molybdenum ($\bar{T}^\beta = 2900^\circ\text{K}$), tungsten ($\bar{T}^\beta = 3650^\circ\text{K}$), rhenium ($\bar{T}^\epsilon = 3450^\circ\text{K}$), ruthenium ($\bar{T}^\epsilon = 2550^\circ\text{K}$), osmium ($\bar{T}^\epsilon = 3300^\circ\text{K}$), rhodium ($\bar{T}^\alpha = 2240^\circ\text{K}$), iridium ($\bar{T}^\alpha = 2750^\circ\text{K}$), platinum ($\bar{T}^\alpha = 2040^\circ\text{K}$) and palladium ($\bar{T}^\alpha = 1840^\circ\text{K}$). With the use of thermodynamic identities of the form

$$\Delta F_A^{\alpha \rightarrow L} \equiv \Delta F_A^{\alpha \rightarrow \beta} + \Delta F_A^{\beta \rightarrow L} \quad (3)$$

all of the required free energy differences can now be defined explicitly. As a consequence, the melting points of the unstable forms can be computed. These are shown for convenience in Table I.

The computed phase diagrams are shown by the heavy lines in Figures 3 to 17. These figures also show currently available experimental phase relations generated in a large measure as a result of recent research at Nuclear Metals Corporation of Concord, Massachusetts (10, 19). The calculations do not consider compound phases. These are shown as cross hatched regions on the phase diagram. As anticipated, the existence of compound phases will limit the range of stability of the β , ϵ , α or L phases. In addition, where compound phases are dominant or numerous, no meaningful comparison of computed and observed

TABLE I
SUMMARY OF COMPUTED MELTING POINTS

<u>Element</u>	<u>$\bar{T}^{\beta} (^{\circ}\text{K})$</u>	<u>$\bar{T}^{\epsilon} (^{\circ}\text{K})$</u>	<u>$\bar{T}^{\alpha} (^{\circ}\text{K})$</u>
Nb	2740	1420	1170
Ta	3270	1800	1540
Mo	2900	1900	1530
W	3650	2650	2230
Re	2710	3450	2890
Ru	1420	2550	1780
Os	1060	3300	2310
Rh	930	2010	2240
Ir	1260	2490	2750
Pd	820	1470	1840
Pt	980	1670	2040

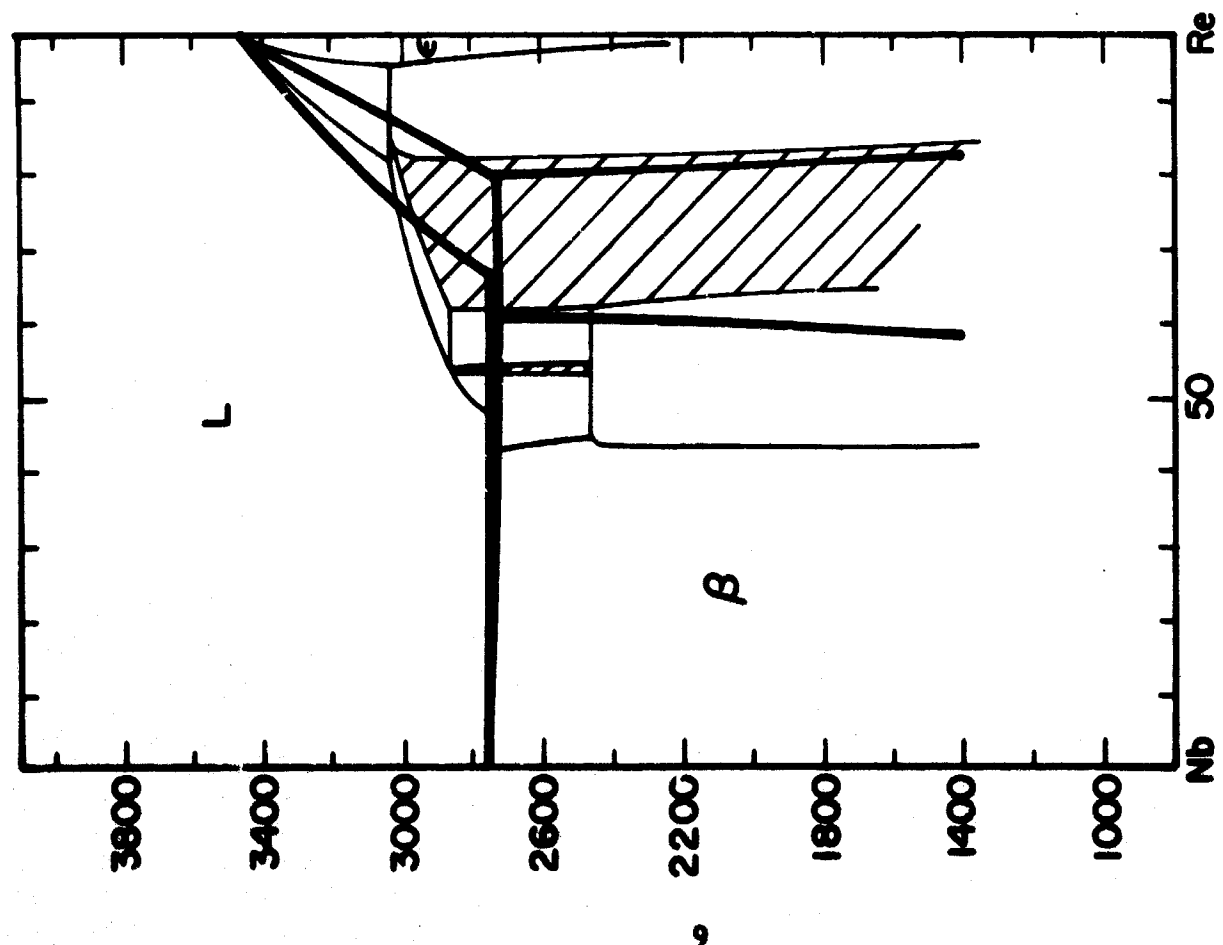
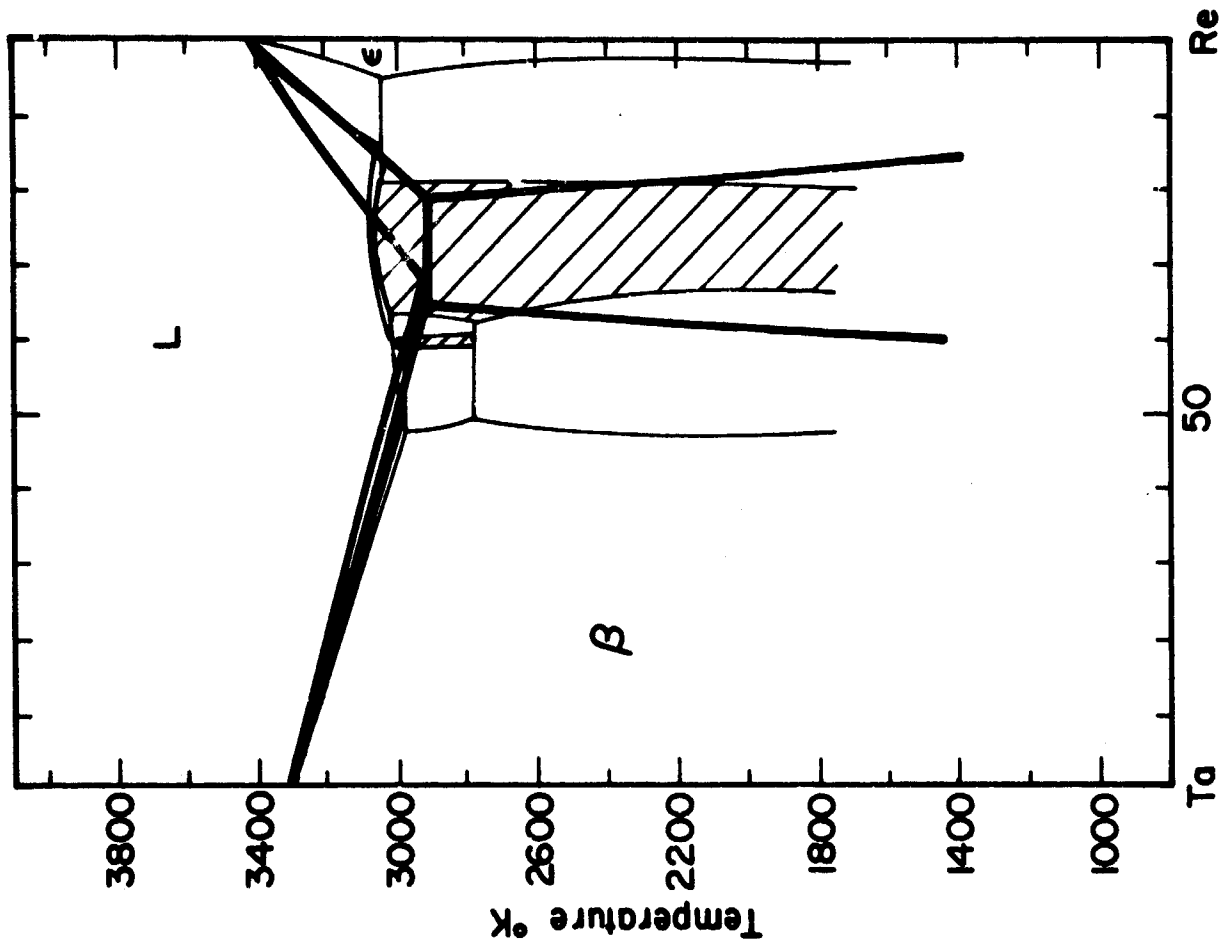


Figure 3. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (7,8).

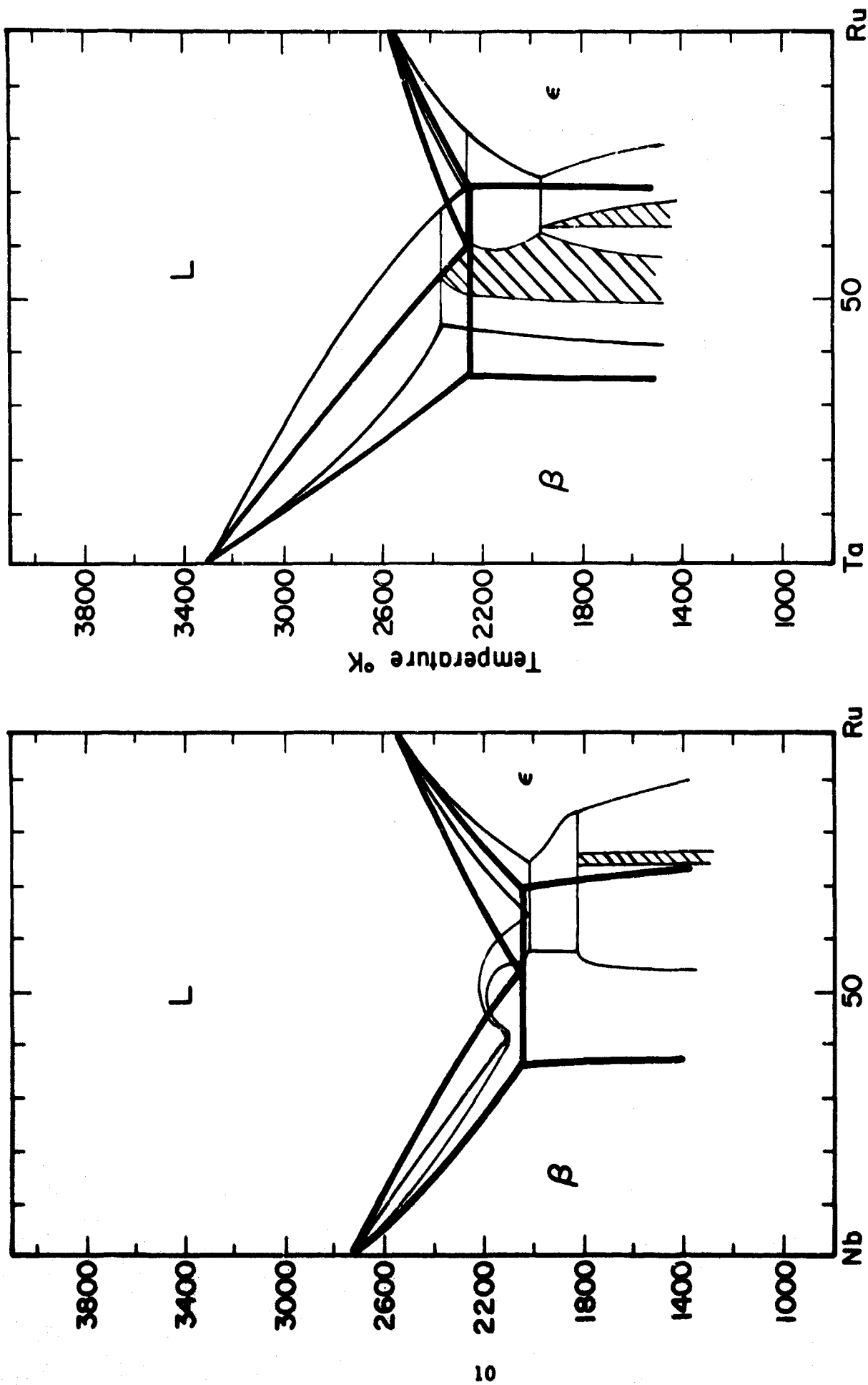


Figure 4. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (9, 10).

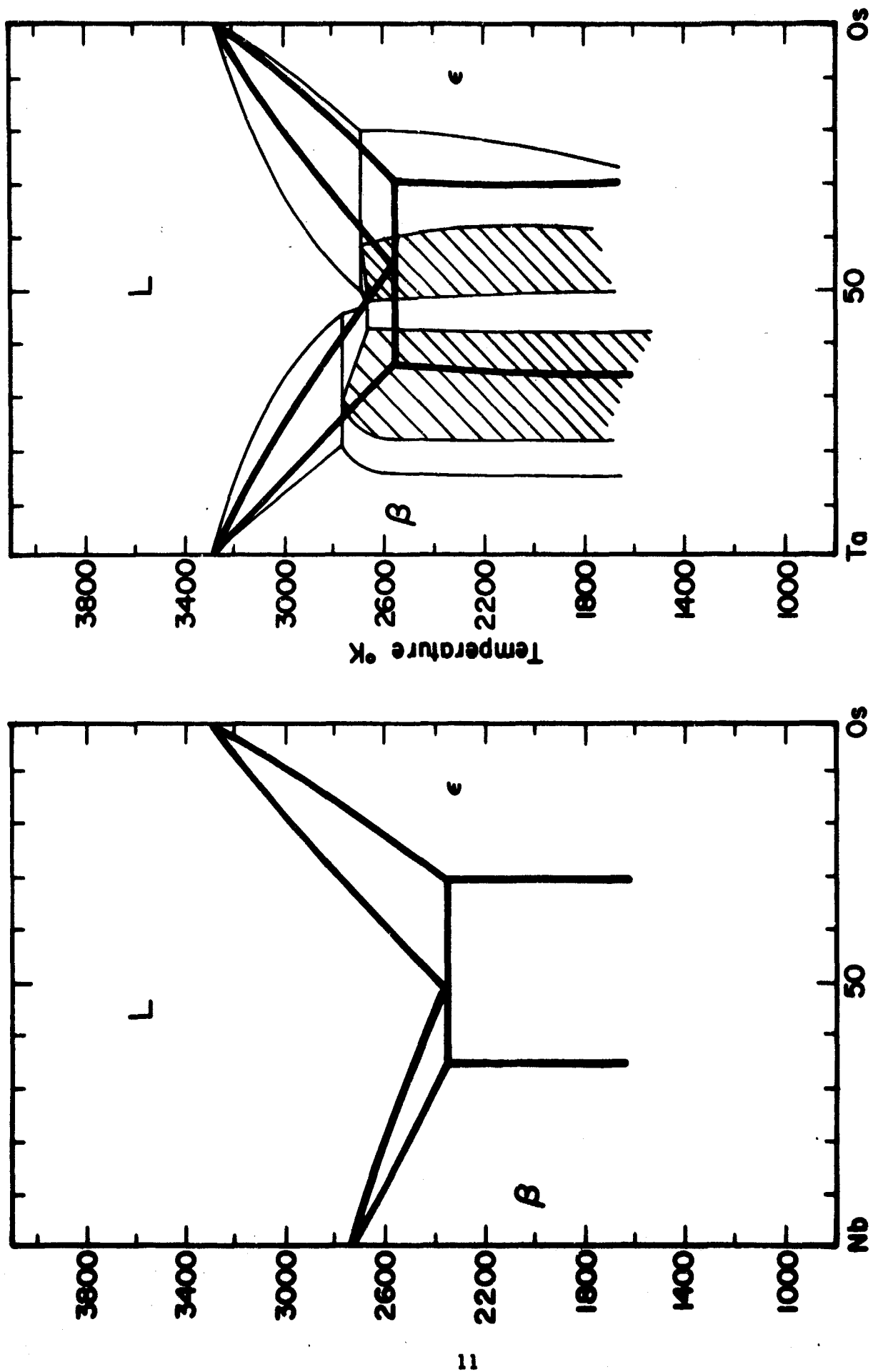


Figure 5. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (19).

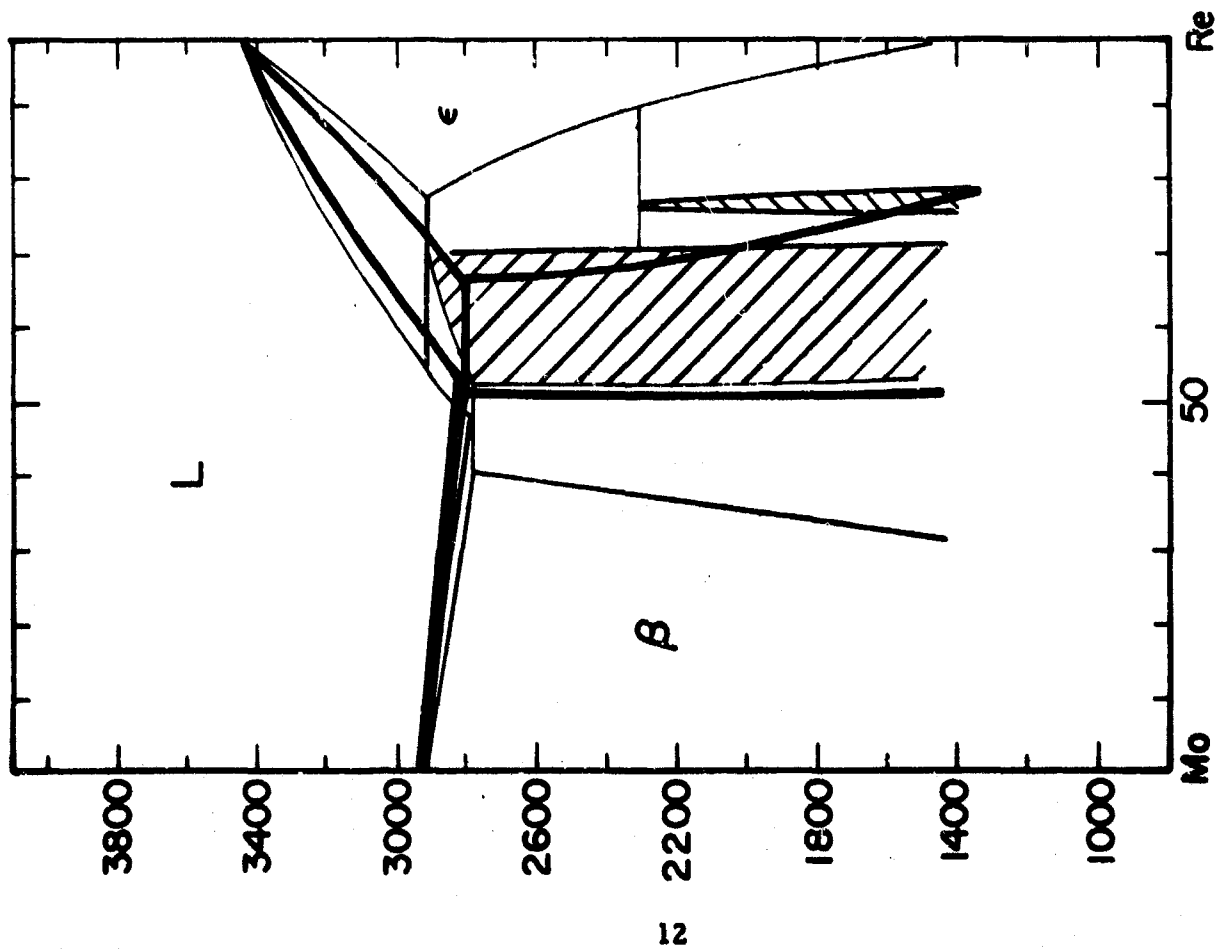
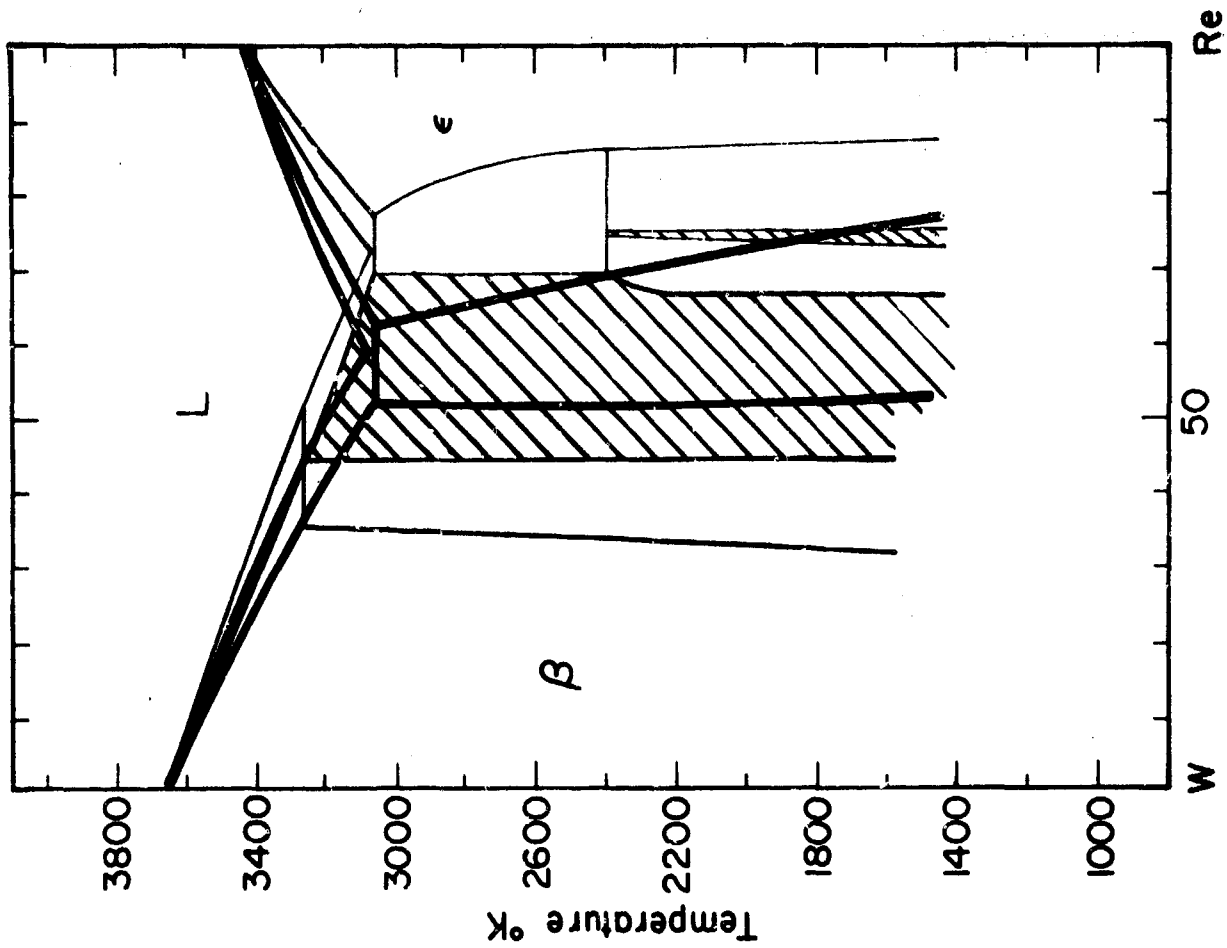


Figure 6. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (11, 12).

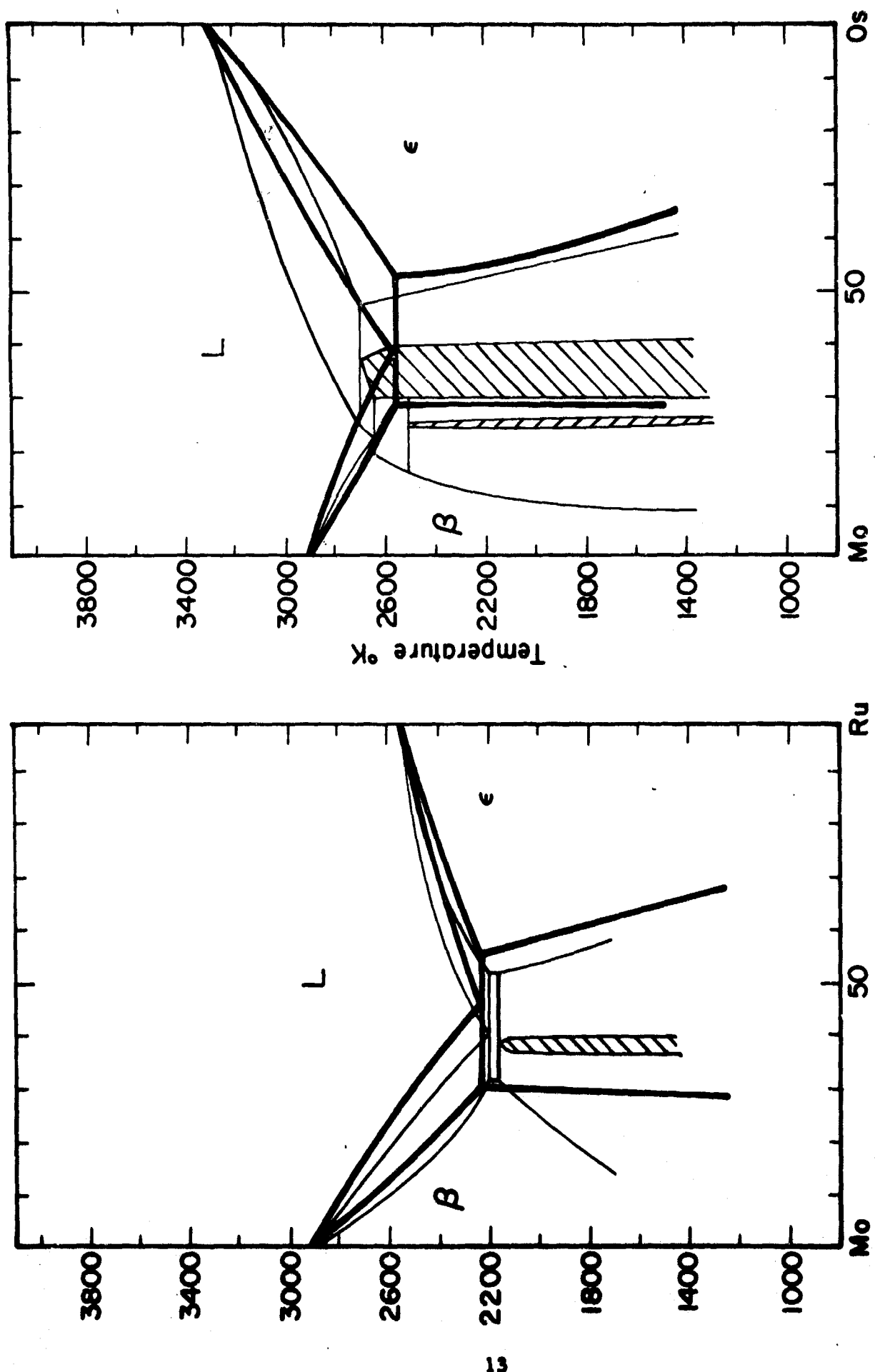


Figure 7. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (13, 14).

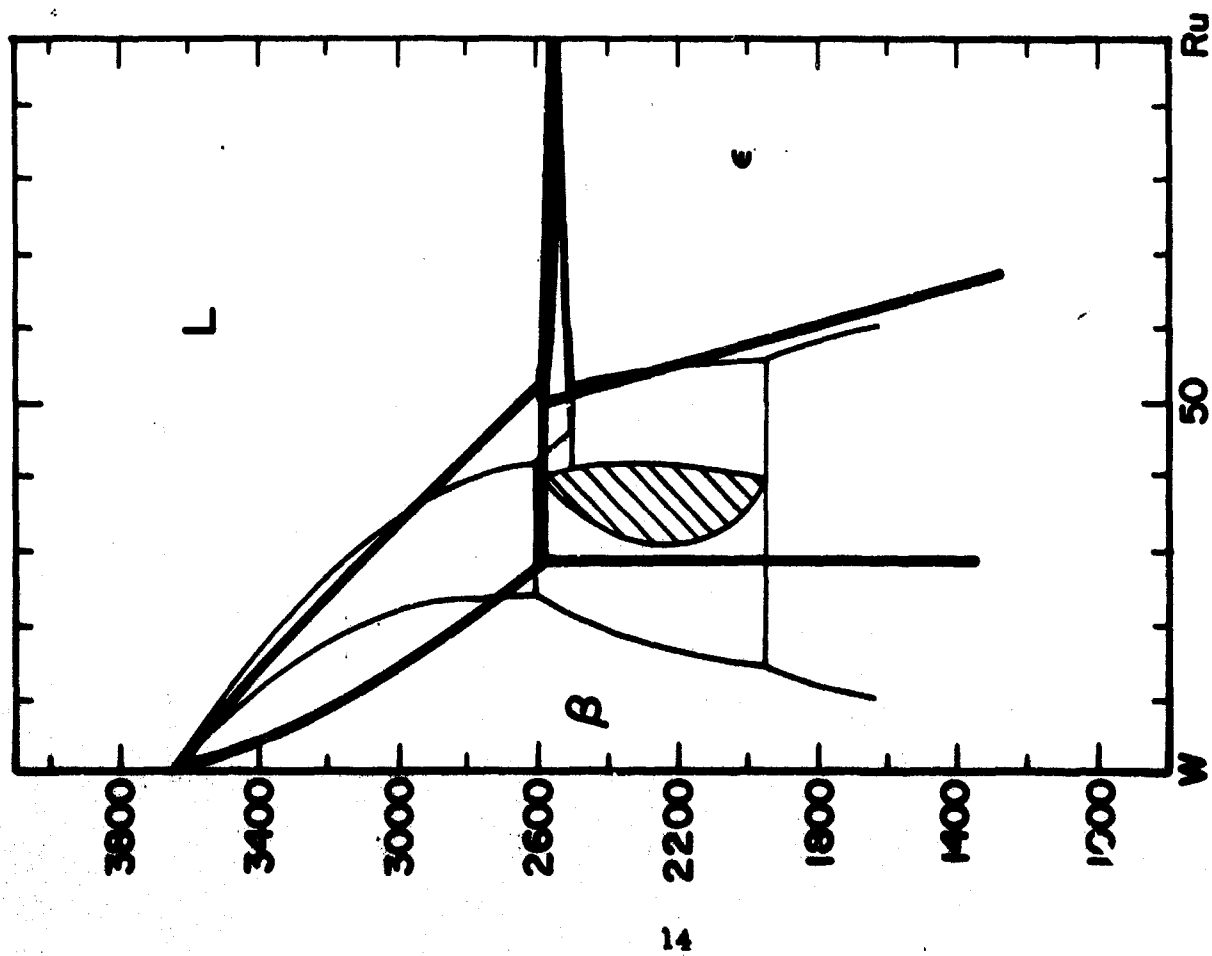
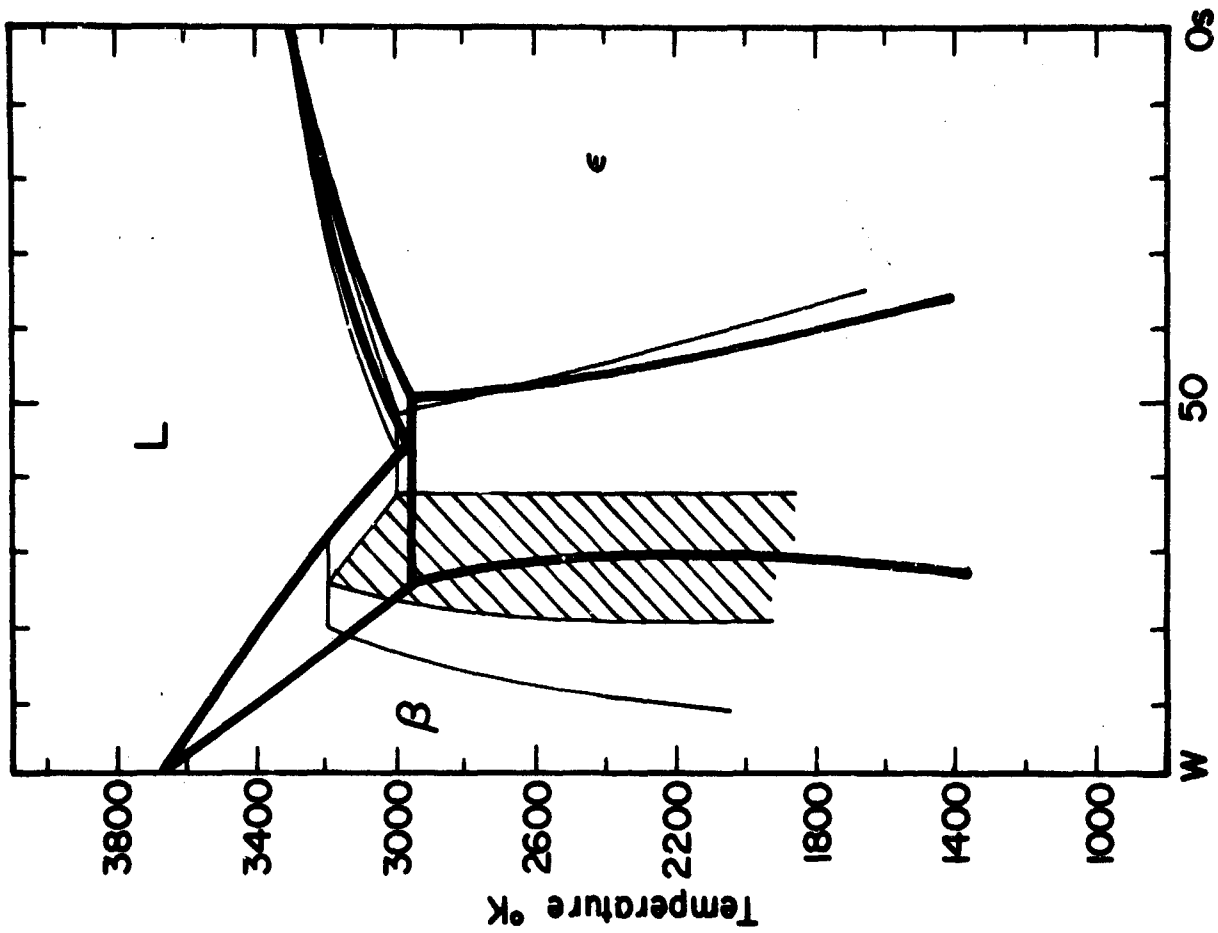


Figure 8. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (15, 16).

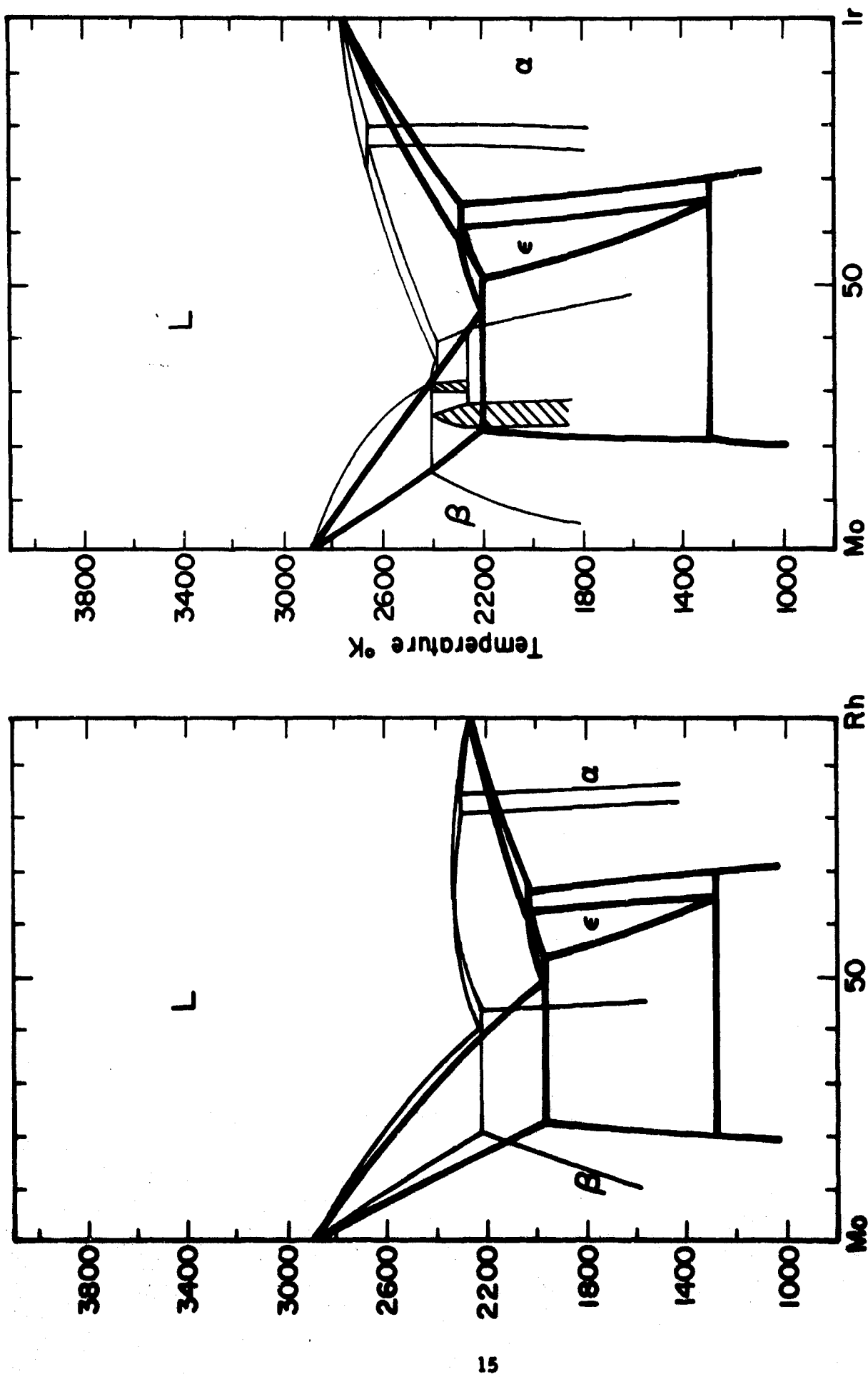


Figure 9. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (17, 18).

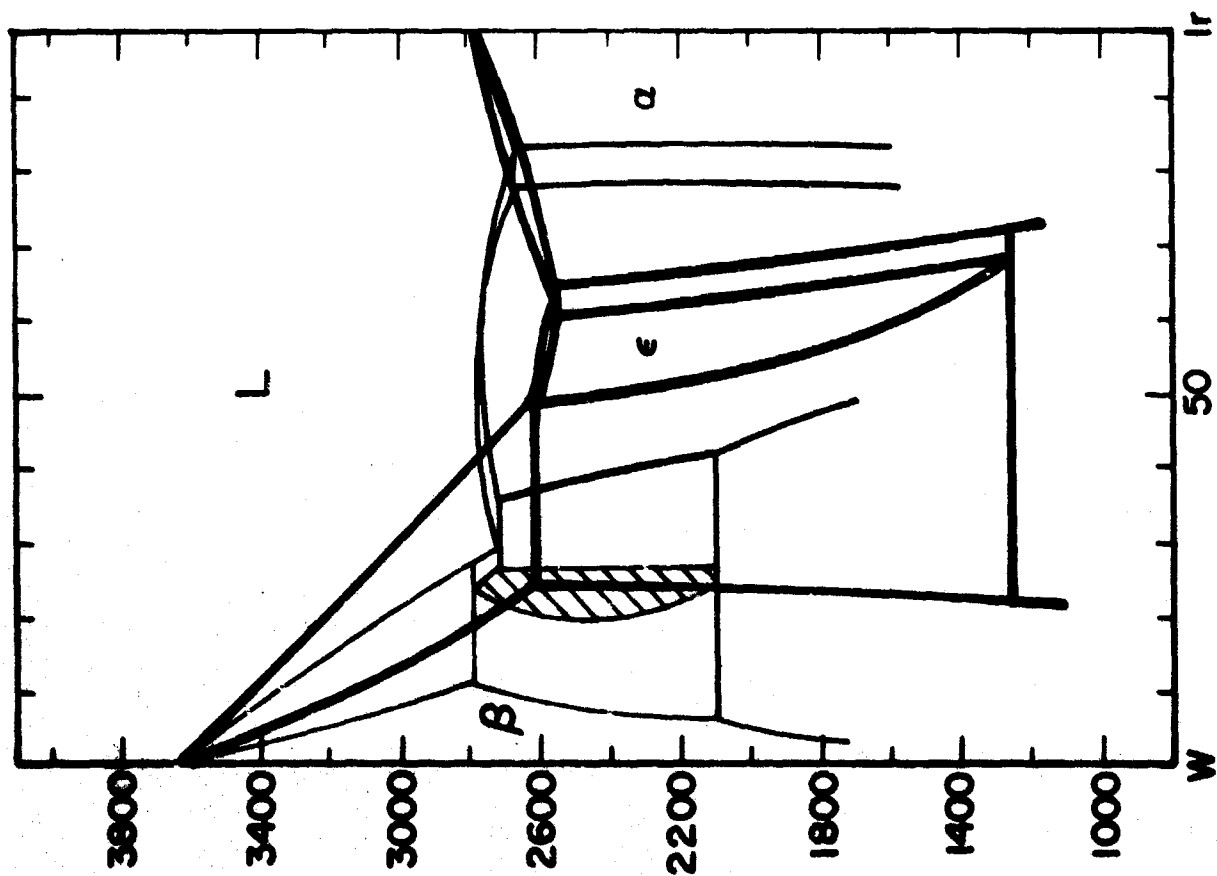
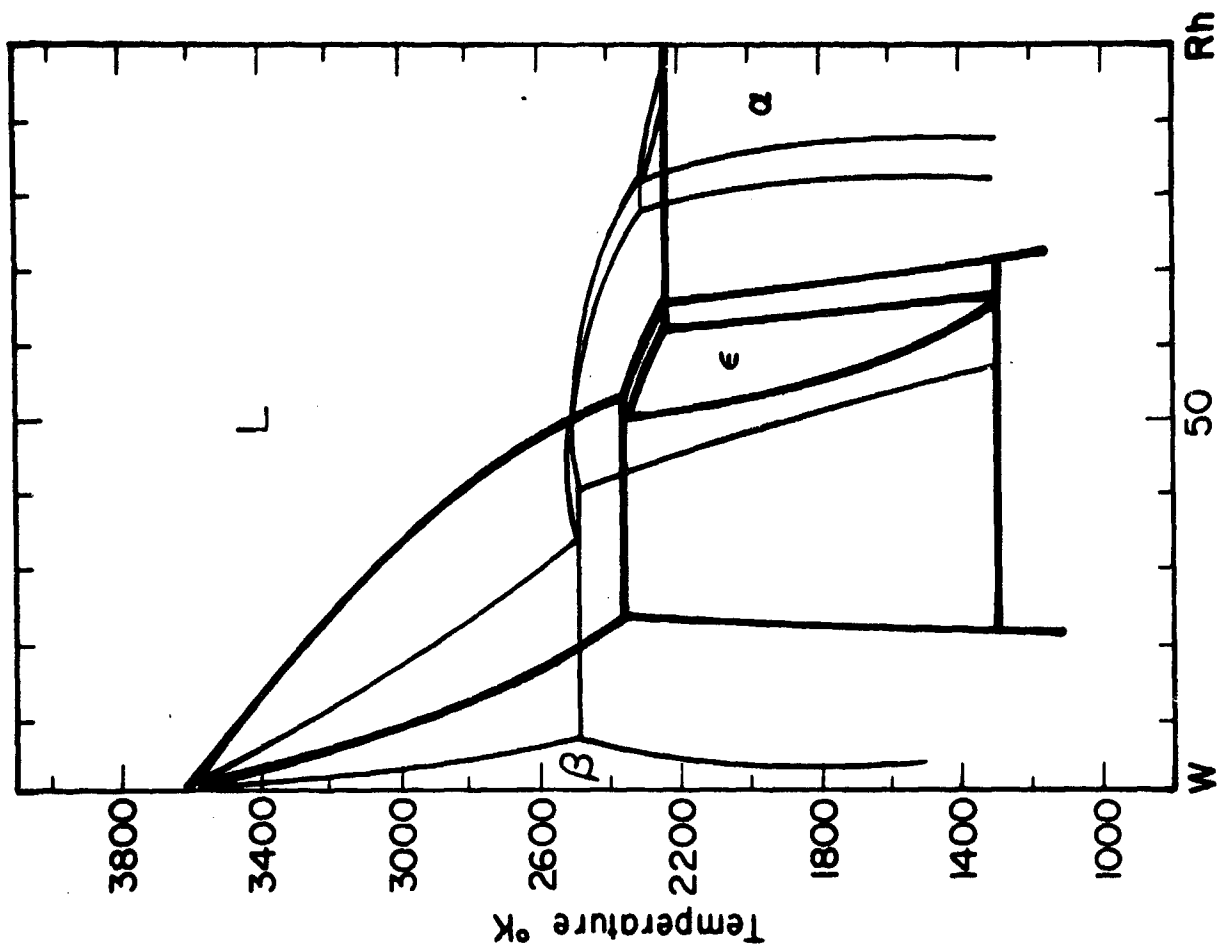


Figure 10. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (19, 19).

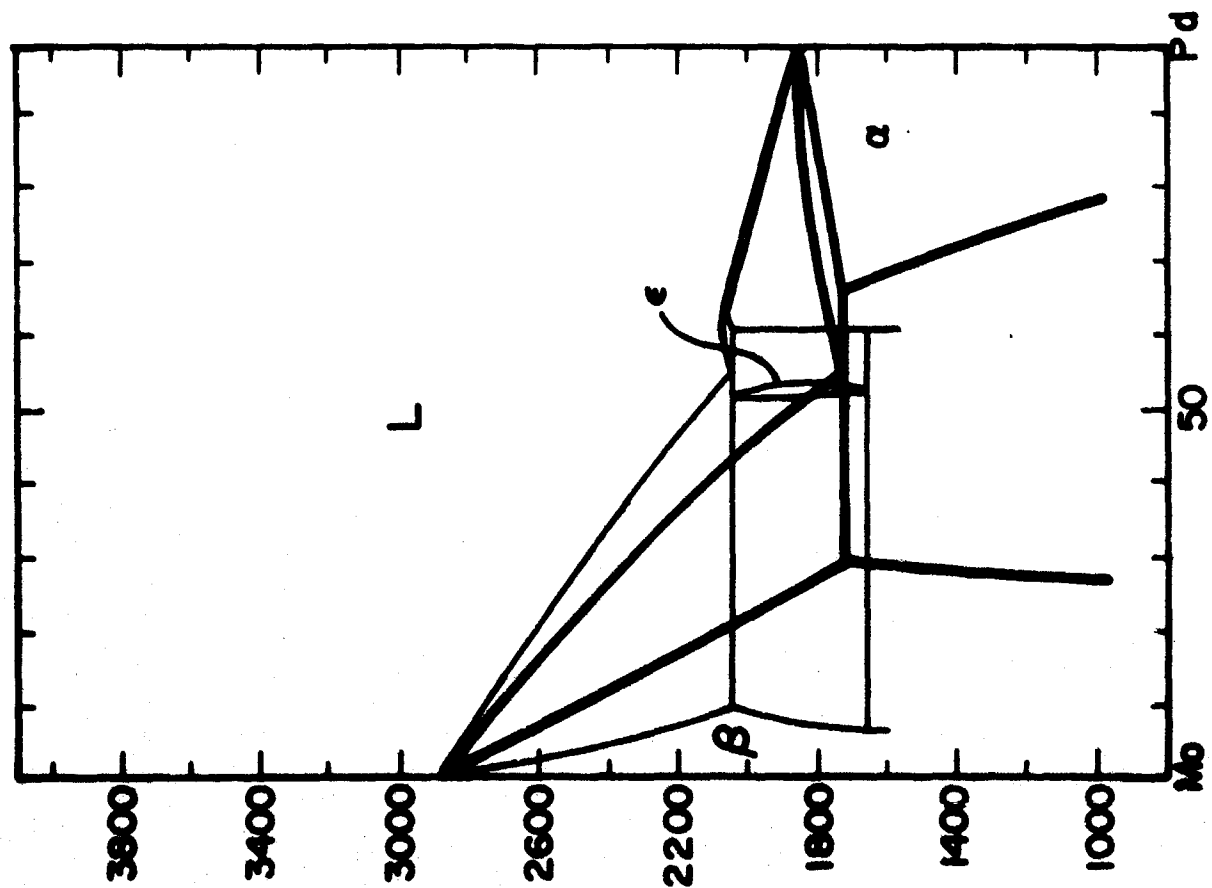
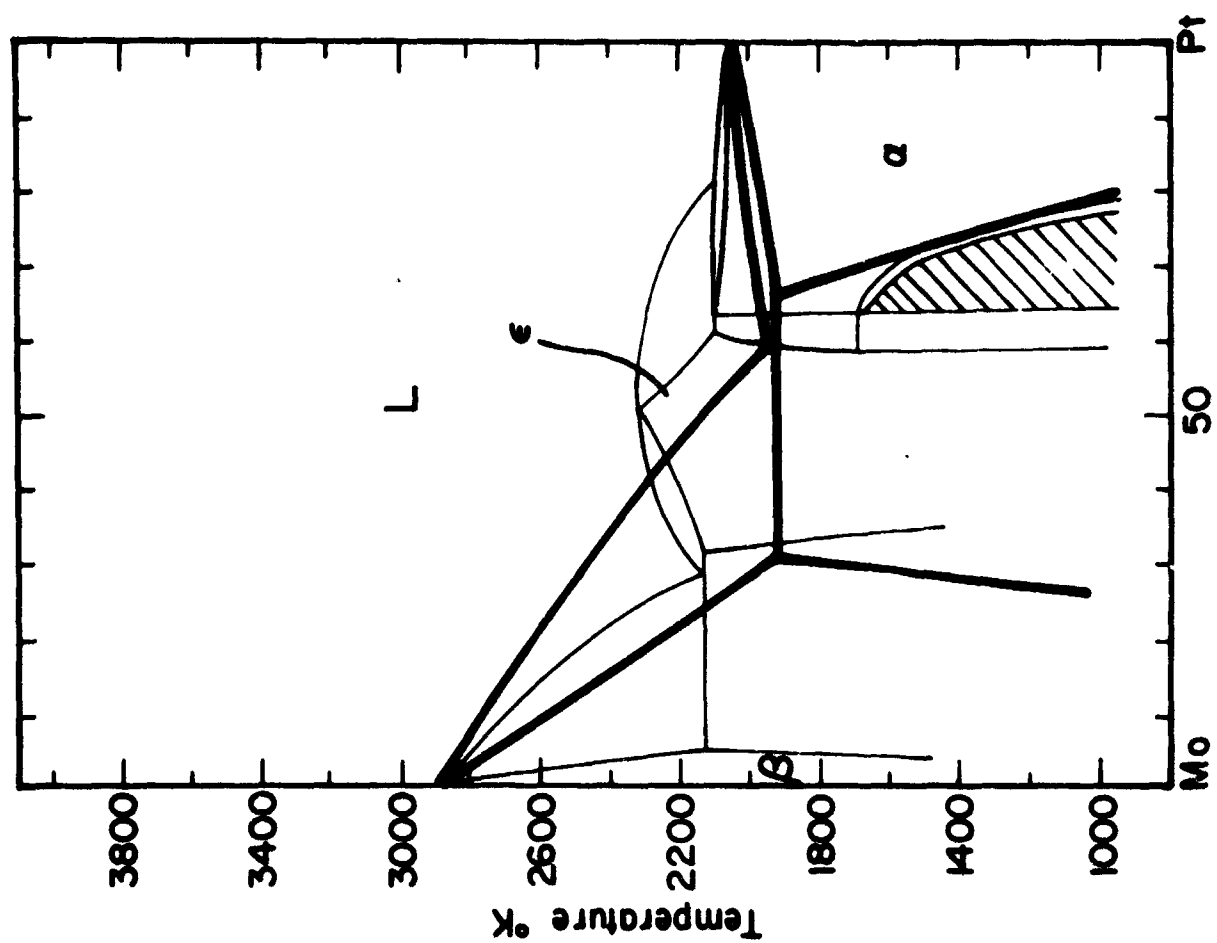


Figure 11. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (20, 21, 22).

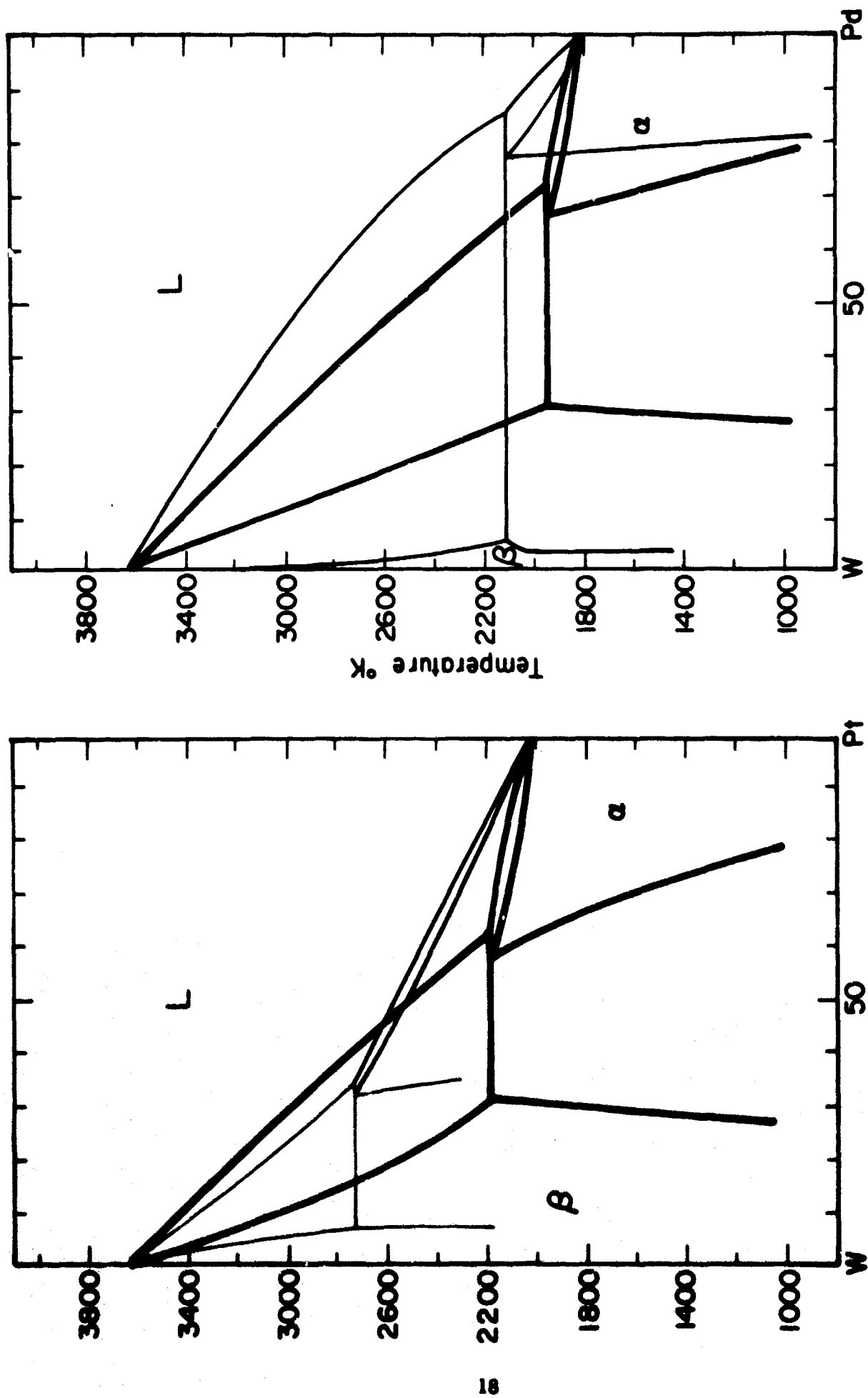


Figure 12. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (23,24).

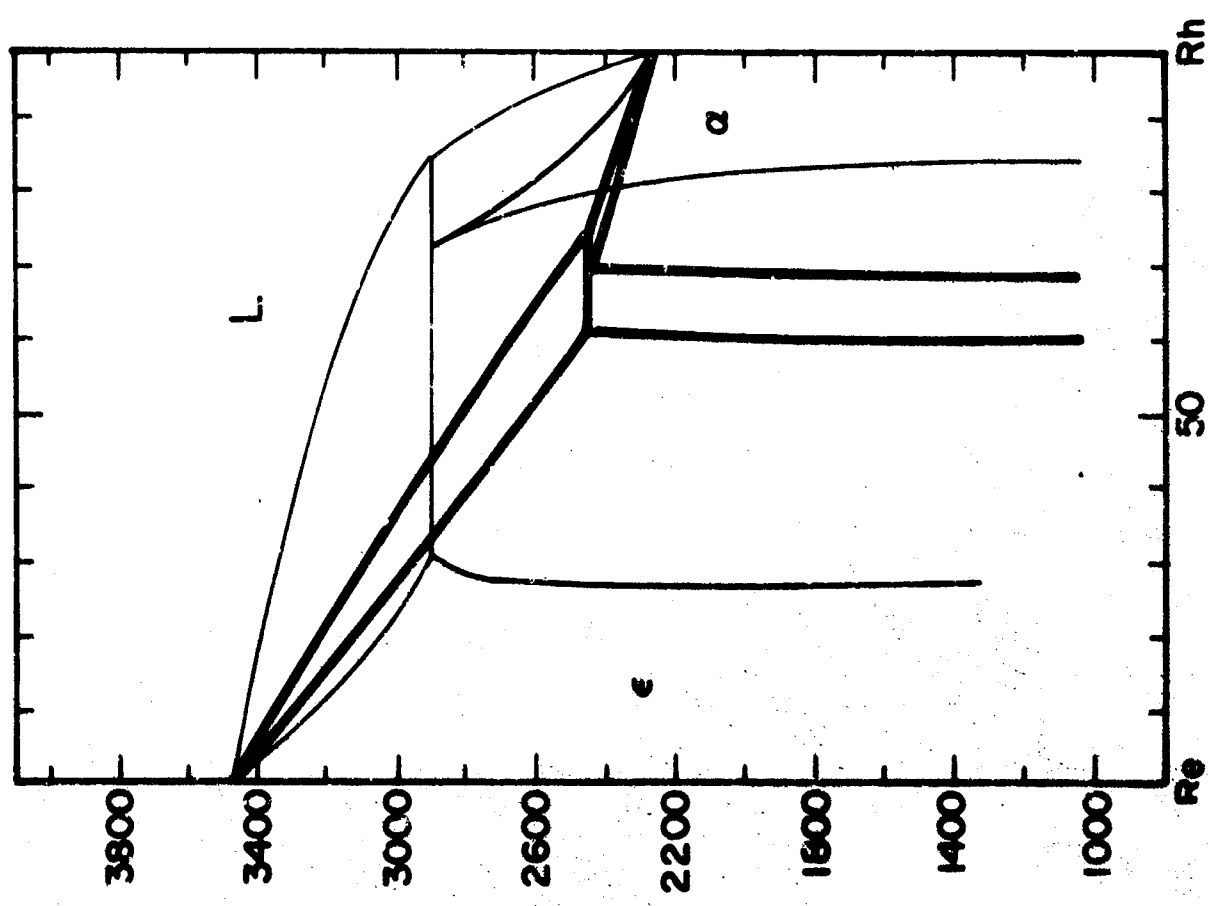
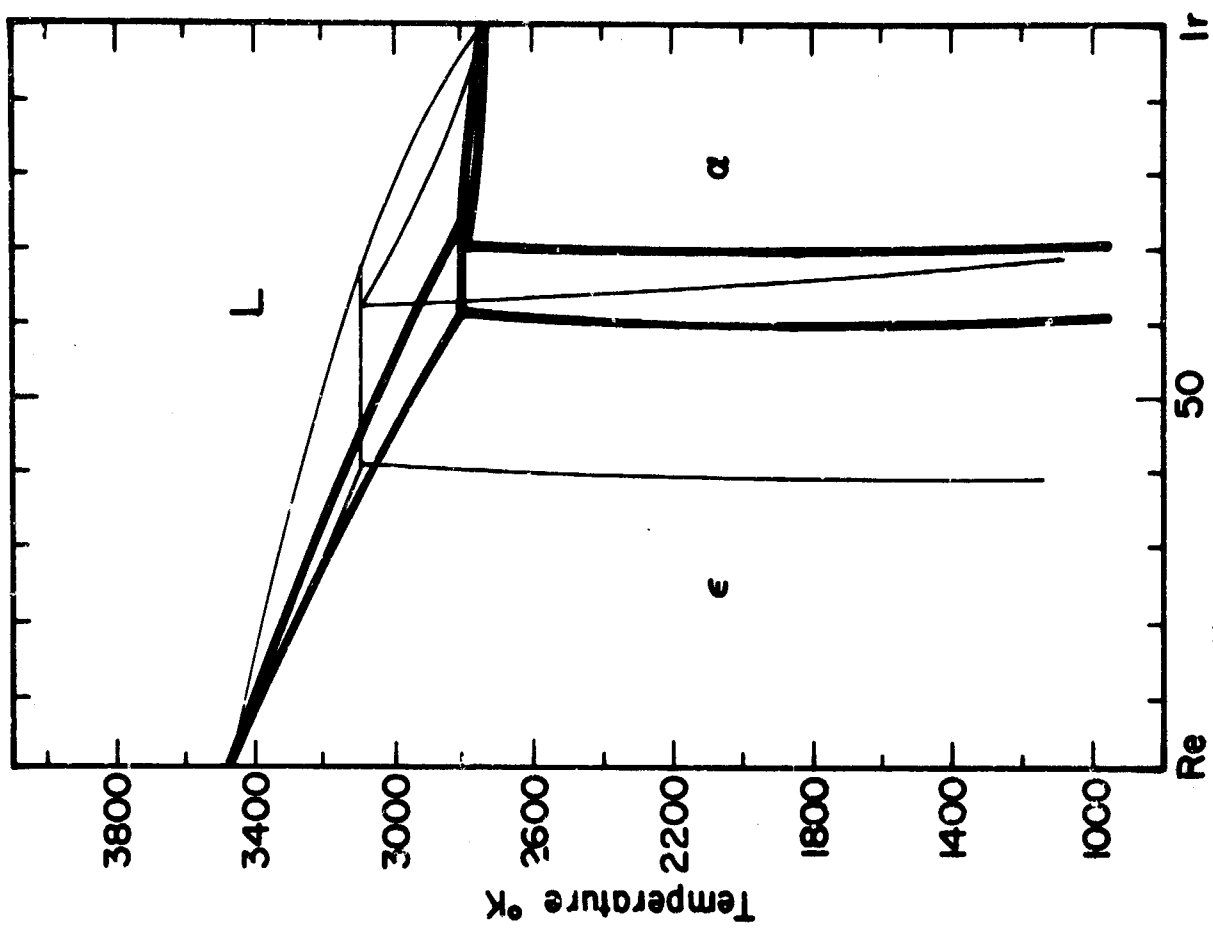


Figure 13. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (25, 25).

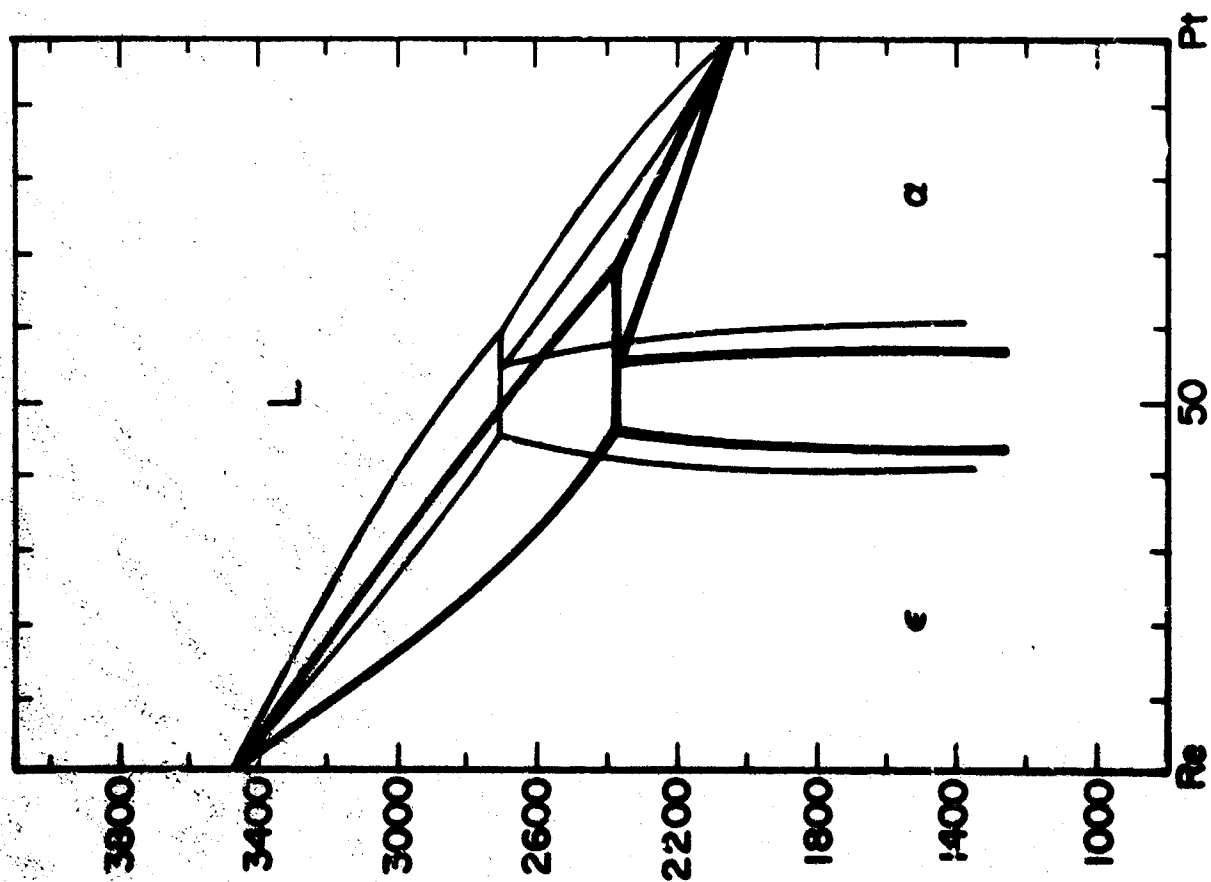
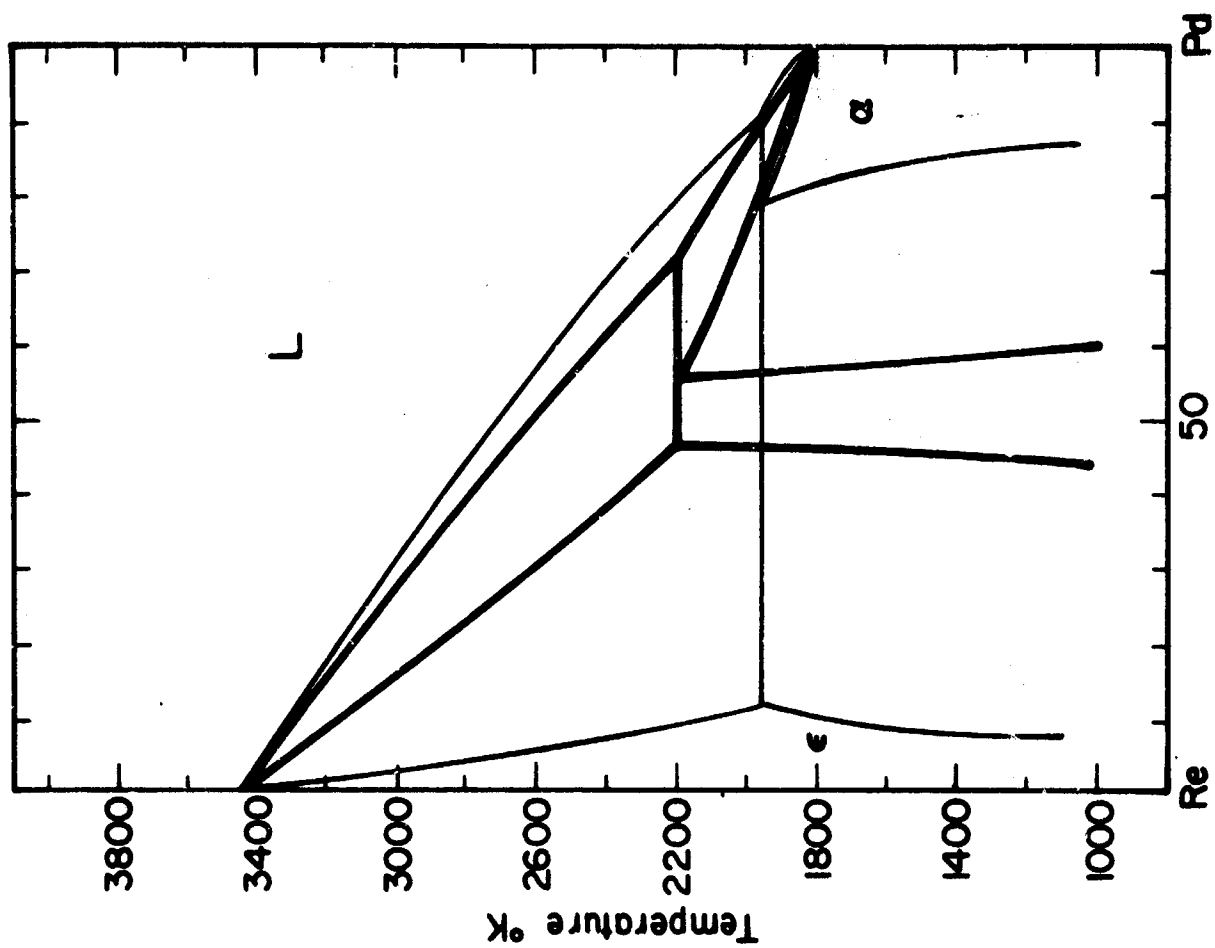


Figure 14. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (26, 25).

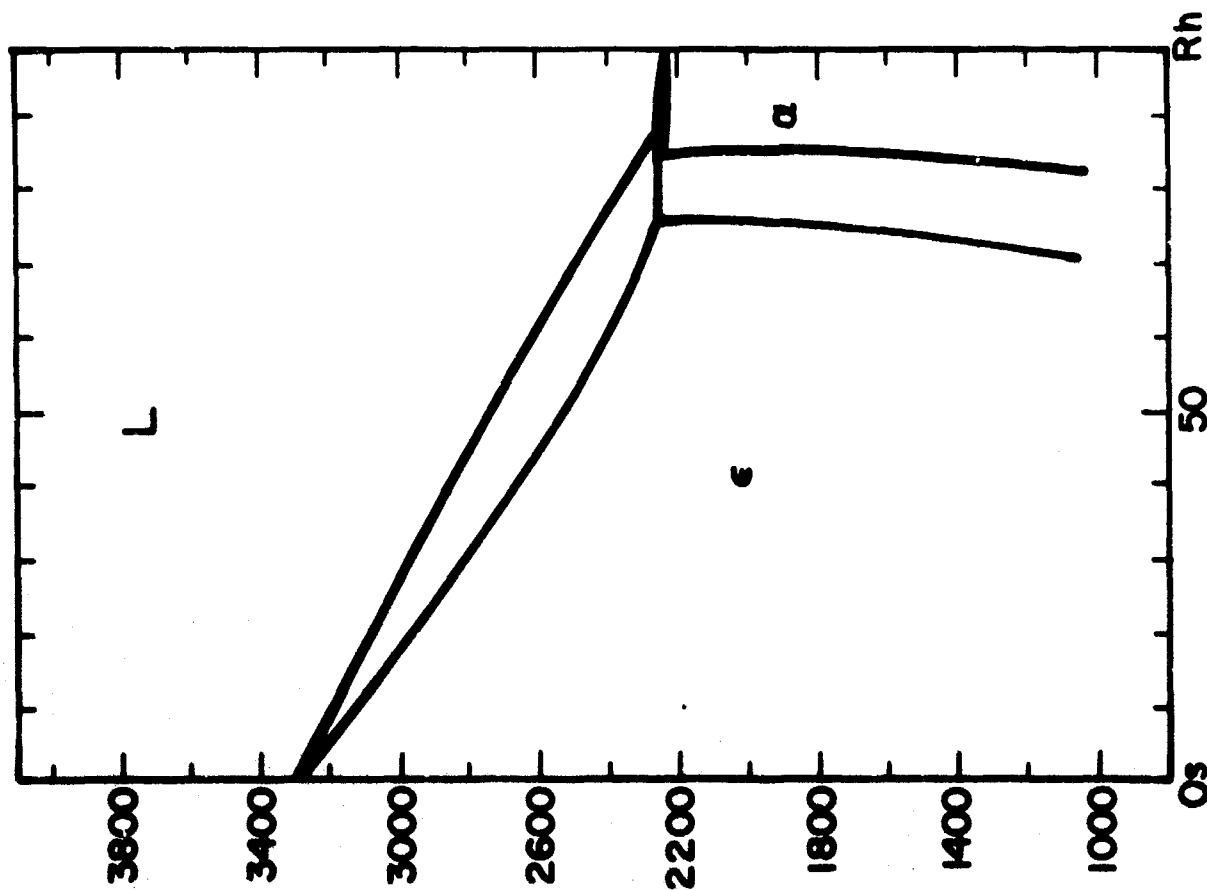
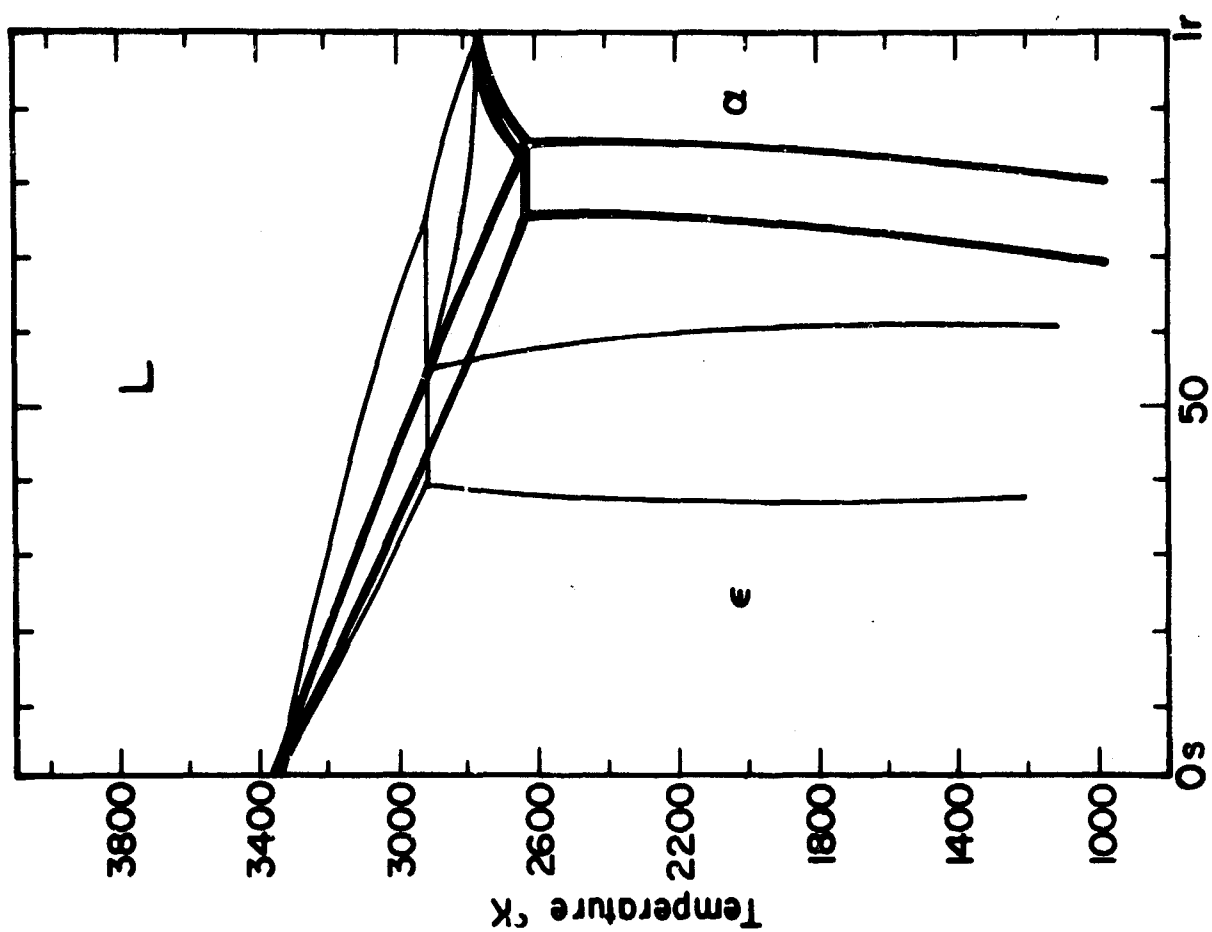


Figure 15. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (27).

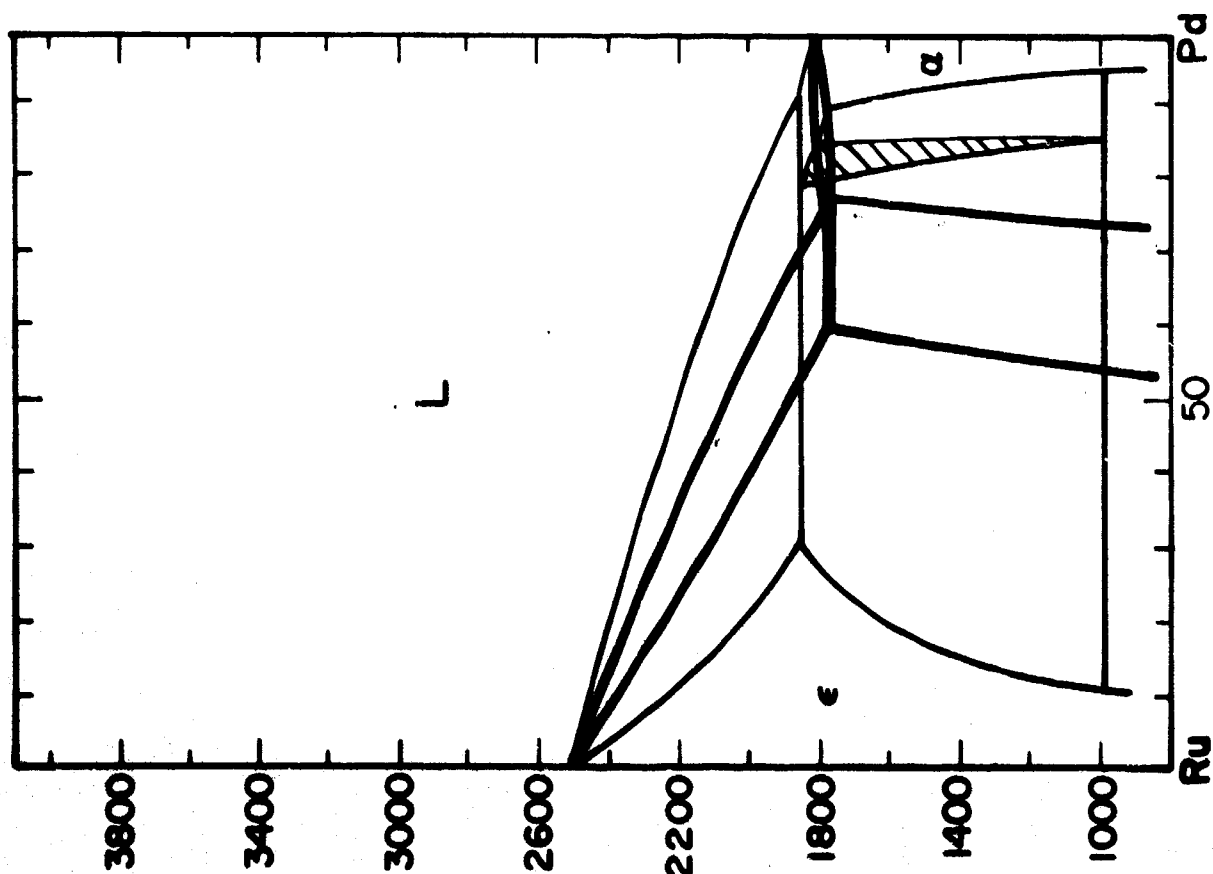
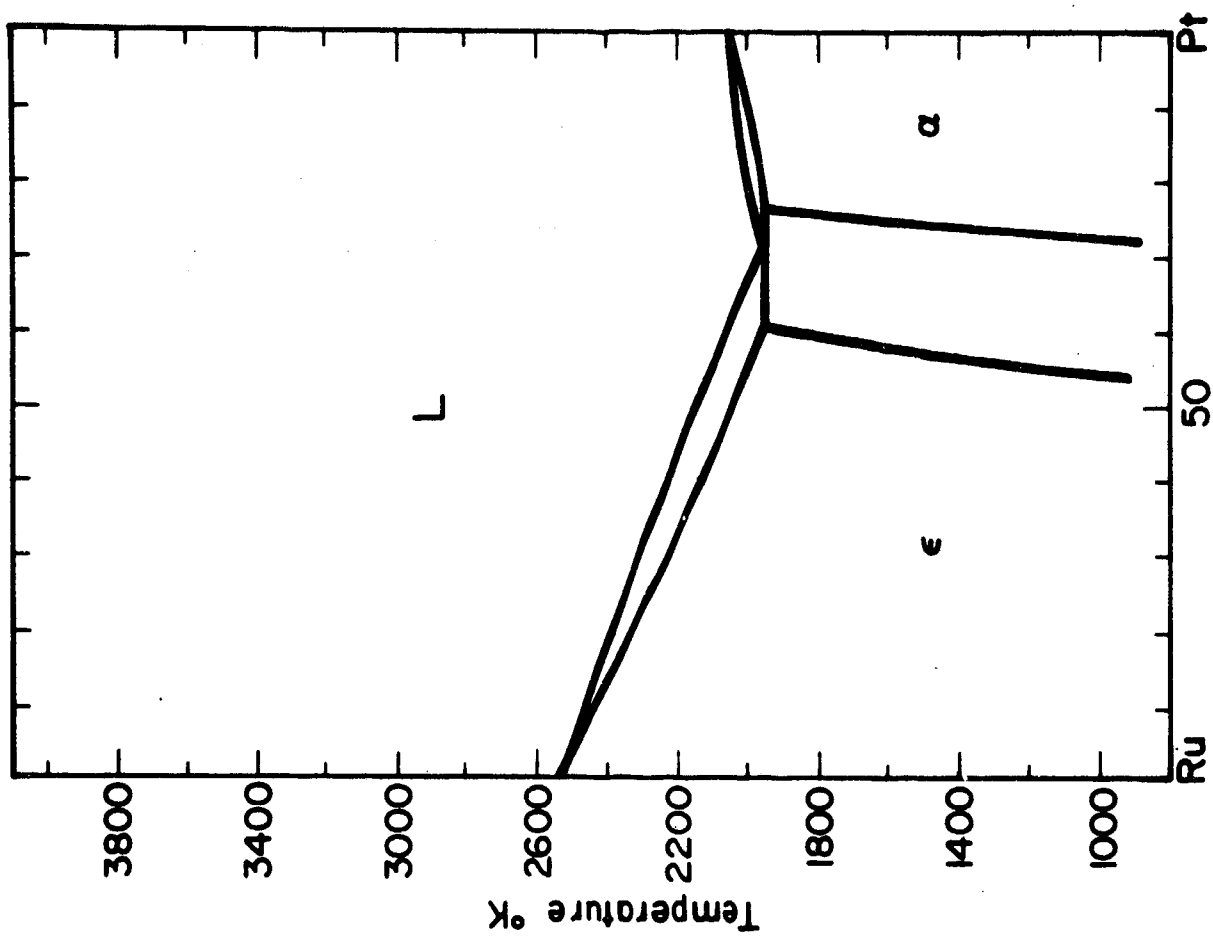


Figure 16. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results (28).

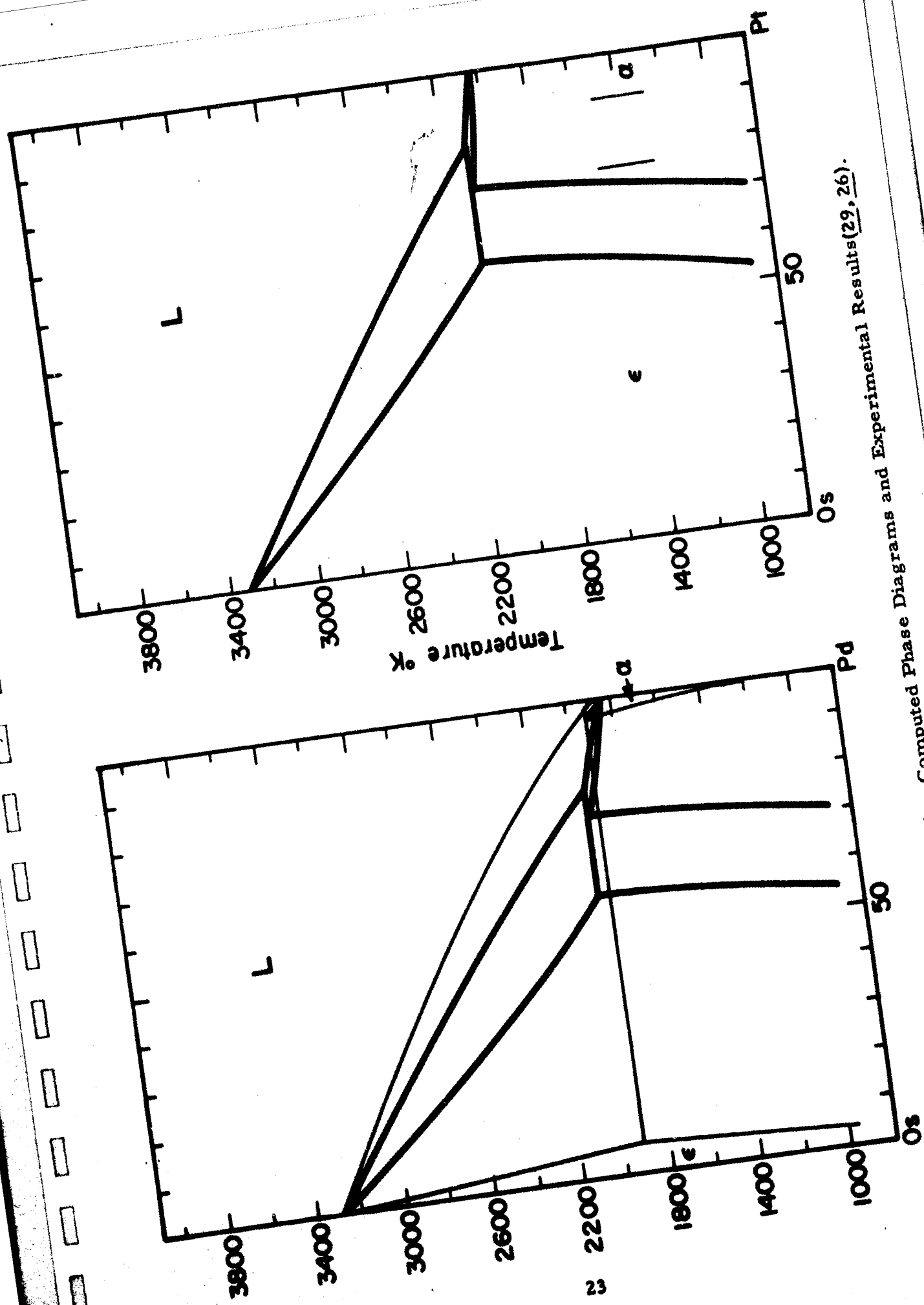


Figure 17. Comparison of Ideal Solution Computed Phase Diagrams and Experimental Results(29, 26).

behavior can be made. As a result, the binary phase diagrams for either Zr or Hf with each of the elements Tc, Re, Ru, Os, Rh, Ir, Pd and Pt are not presented. Similarly the binary diagrams for either Nb or Ta with each of the elements Rh, Ir, Pd and Pt are not considered. Finally, in the binary systems between Zr or Hf with each of the elements Nb, Ta, Mo and W, miscibility gaps dominate the binary systems. These cases will be treated at a later date when the ideal solution approximation* (i.e., $A = B = E = L = 0$) is relaxed. Finally, the Tc and Re group is represented by rhenium due to the paucity of information concerning technetium. These diagrams can be easily computed for comparison with observed phase equilibria when the latter are available.

Figure 3 illustrates the intrusion of compound phases. However, the $\beta/\epsilon/L$ equilibria are in good accord with observation. This is equally true in Figure 4 although the β/ϵ equilibrium in the Nb-Ru system is computed to be too near niobium. This is probably due to the fact that deviations in the β phase are more negative than ϵ (i.e., $B < E$). Figure 5 is similar to Figures 3 and 4 showing the compound effect; nevertheless the $\beta/L/\epsilon$ equilibria are in agreement with the computations. The tungsten-rhenium and molybdenum-rhenium cases shown in Figure 6 are similar to those cited above as are the Mo-Ru and Mo-Os examples in Figure 7. In the latter case, the β/ϵ equilibria are shifted too far toward osmium (i.e., E is probably less than B). Good agreement is found in Figure 8 for the W-Ru and W-Os cases. Figures 9 and 10 show the interesting case where the calculations actually predict the stability of the h.c.p., ϵ , phase. The larger range of stability of the ϵ phase observed is probably due to negative

* The regular solution parameters E and L are defined in Eqs. (1) and (2). The parameters A and B are corresponding terms for the α and β phases.

deviations (i.e., $E < 0$ and $E < A, B$, and L). In the Mo-Pt, Mo-Pd, W-Pt and W-Pd cases the h.c.p. phase is almost, but not quite, stable according to the calculations. Interestingly ϵ appears in the Mo-Pt case (Figure 11) but not in the tungsten binary systems. The α/β equilibria in W-Pt are shifted farther toward the tungsten rich side due to negative deviations in the α phase ($A \leq 0$ and $A \leq B, L$). The latter condition would also account for the low computed peritectic. Similarly, the width of the two phase $\beta+L$ and $\alpha+\beta$ fields in the W-Pd are probably due to deviations from ideality. Similar deviations are also seen in Figure 13.

The rhenium-platinum case shown in Figure 14 is in good agreement with observation although in this case E is probably equal to A and both are less than L . A similar situation prevails in the Re-Pd case where positive values of the interaction terms are indicated by the width of the two phase fields.

The computed Os-Ir phase diagram shown in Figure 15 illustrates the poorest agreement between calculated and observed behavior. The difference could be due to the fact that A is less than E and L or that the magnitude of the entropy difference between the α and ϵ phases previously computed for the Ru/Os case (6) is too large (see Figure 2). The Ru-Pt, Ru-Pd, Os-Pt and Os-Pd series shown in Figures 16 and 17 indicate positive deviations in the palladium systems and only sketchy phase diagram data in the case of the platinum systems. However, the Os-Pt deviation is in the opposite direction from the Os-Ir case. Thus, if both systems were ideal, the Os-Pt case would demand larger values for $\Delta F_{Os}^{\epsilon \rightarrow \alpha}$ as opposed to the smaller values required by the Os-Ir deviation. As a result, the lattice stability parameters for Ru/Os shown in Figure 2 are probably correct. The discrepancies shown in the Os-Ir and Os-Pt cases can be rectified by having $A < E$ in the Os-Ir case and $E < A$ in the Os-Pt case.

III. CONCLUSIONS

The broad definition of lattice stability presented here is grossly oversimplified. In addition, the ideal solution constraint used in computing these diagrams is but a crude first approximation. Provisions are currently being made to reduce the phase boundary calculations to computer format so that regular solution phase diagrams can be readily generated in order to re-examine the cases treated here in detail and to incorporate those systems dominated by miscibility gaps.

Nevertheless, the surprising level of agreement between the observed and computed phase diagrams indicates that major revisions of the current lattice stability parameters will not be required.

IV. ACKNOWLEDGEMENT

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<p>The enthalpy and entropy differences between the b.c.c., h.c.p. and f.c.c. structures have been estimated for transition elements of the second and third periods. Continuous curves depicting these differences as a function of electron number in the zirconium through cadmium and hafnium through gold series are presented. The difference parameters have been used to compute thirty binary phase diagrams between elements in the niobium/tantalum through palladium/platinum groups. These binary diagrams, which are restricted to cases where the end members exhibit differing stable crystal structures and miscibility gaps are absent, have been calculated on the basis of ideal liquid, b.c.c., h.c.p. and f.c.c. phases. The latter simplification has been chosen in order to afford the clearest possible comparison of computed and observed phase diagrams.</p>		

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